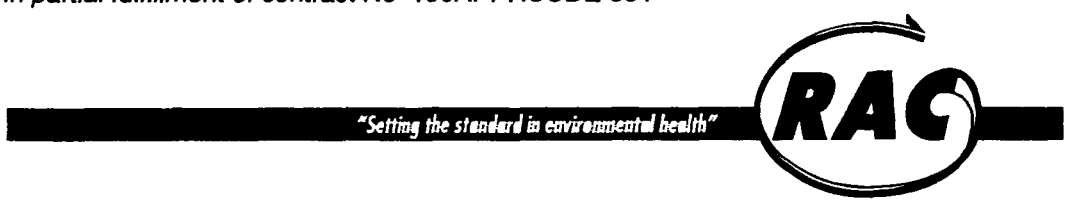
DRAFT REPORT

Estimated Exposure and Cancer Risk from Beryllium Released to the Air from the Rocky Flats Plant

Part of Task 3· Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals

February 1997

Submitted to the Colorado Department of Public Health and Environment Disease Control and Environmental Epidemiology Division Rocky Flats Health Studies in partial fulfillment of contract No 100APPRCODE 391



Radiological Assessments Corporation
417 Till Road Neeses South Carolina 29107
phone 803.536.4883 fax 803.534 199

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Part of Task 3: **Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals**

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Authors

Patricia D McGavran, Ph.D , McGavran Toxicology Consulting, Inc
Arthur S Rood, K-spar, Inc.

Principal Investigator

John E Till, Ph.D , *Radiological Assessments Corporation*

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INTRODUCTION

This report documents risk calculations for inhalation of beryllium in air resulting from normal operational releases at the Rocky Flats Plant (RFP) A brief review of the Phase I work and beryllium source terms is provided It evaluates soil and sediment monitoring data for beryllium and discusses regulatory guidelines, evidence of carcinogenicity and chronic beryllium disease The report also describes environmental transport modeling, estimates of uncertainty in the model predictions and presents distributions of carcinogenic risk resulting from inhalation of beryllium for several generic receptor scenarios

BERYLLIUM SOURCE TERM ESTIMATES

Beryllium Use at the Rocky Flats Plant

Although, initial research and developmental work with beryllium began in 1953 at the Rocky Flats Plant, foundry operations with beryllium became significant from 1958 to 1975 Details of beryllium component manufacturing, machining, cutting, heat treating, rolling and other operations and ventilation systems used to control beryllium emissions over the years are described in the Task 3 & 4 Report for Phase I of the Rocky Flats Dose Reconstruction Project (ChemRisk 1992) and in a letter written by Campbell (1986) Most of the beryllium work was done in Building 444 and Building 883 The airborne emission points for beryllium are listed in Table 5-1, pages 169–176 of the Phase I Task 3&4 Report (ChemRisk 1992) With the possible exception of effluent from Building 441 in the early 1960s, all air exhaust discharged from plant facilities that processed beryllium was subjected to high-efficiency particulate air (HEPA) filtration designed for controlling radioactive effluents (ChemRisk 1994a)

Beryllium Releases Estimated from Effluent Monitoring Data

Beryllium has been monitored in the plant air exhaust effluent since at least 1963 (ChemRisk 1992, 1994a, Hammond 1963) The Phase I Task 5 Report describes the monitoring program and summarizes the release data generated as a result of the monitoring program (ChemRisk 1994a)

The monitoring program data for routine airborne emissions of beryllium served as the basis for the Phase I release estimates (Tables 1 and 2, and Figure 1) ChemRisk compiled a record of beryllium emissions using sample data logbooks for 1960 through 1970 and annual beryllium releases reported in the annual Environmental Monitoring Reports for 1971 through 1989 The logbooks contain daily sample results for workroom air and building effluents The monthly and annual average beryllium concentrations for each stack were calculated from the building effluent data Data on exhaust flow rates and total exhaust volume were lacking for some facilities and had to be estimated using facilities of similar size No sampling data from before 1960 were located It was assumed that emissions in 1958 and 1959 were the same as those reported in 1960

Air exhaust samples were taken from filter plenum exhausts after the air passed through HEPA filters but before it exited the stack The sampling practices, sampling system design sample line losses calculations of flow rates and exhaust volume and uncertainties discussed previously for radioactive radioactive particles (ChemRisk 1994a) were applied to

the beryllium sampling data. The Task 5 Report for Phase I describes the different analytical techniques used over time (ChemRisk 1994a)

The Phase I report (ChemRisk 1994a) also discusses beryllium released during three fires that occurred in Building 444 in 1962, 1964, and 1978 (West 1978, Werkema 1978a) Any releases as a result of the fires would have been monitored by the stack sampling equipment so they were thought to be included in the Phase I release estimates (ChemRisk 1994a) The most significant fire occurred on February 23, 1978 A release estimate of 14 5 g from the fire was incorporated into the Phase I release estimate of <17 g total for 1978. This estimate was based on monitoring results from the plenum sampler, ambient air sampling and water samples from water used to fight the fire that drained into and was sampled from ponds, ditches and temporary impoundments (Rockwell 1978, ChemRisk 1994a, Hawes 1978, Werkema 1978b) Other estimates of a maximum beryllium source term from the fire were 10 kg (Rockwell 1978), and 8-10 kg (West 1978, Werkema 1978a) These release estimates were used to calculate a maximum air concentration of 8 to 10 $\mu g\ m^{-3}$ at Highway 93, using Gaussian plume calculations One calculation led to an estimate of 91.7 kg of beryllium (West 1978), which was later recognized as "so gross an overestimate" as to be discounted (Werkema 1978a)

Beryllium release summaries in the Task 5 report (ChemRisk 1994a) suggest low release values that average tens of grams or less annually Documentation suggests that beryllium measurement data handling practices may have led to reporting annual emissions that were greater than actual releases (ChemRisk 1994a)

Table 1 Total Annual Release Estimates for Beryllium from Table 3-2 of the Phase I Task 5 Report (ChemRisk 1994a)

Year	Annual release estimate (g)	Year	Annual release estimate (g)
1958	13 [a]	1965	31
1959	13 [a]	1966	33
1960	13	1967	33
1961	11	1968	38
1962	6 7	1969	24
1963	12	1970	14
1964	12	Total (1959–1970)	253 7

[a] No monitoring data were found for 1958 and 1959 and releases in those years were assumed to be the same as 1960

Beryllium releases from 1971 to 1989 were obtained from the Annual Environmental Monitoring Reports issued by the RFP These reports often reported beryllium release totals for the year as less-than values The 1975 report explained that samples with concentrations less than the minimum detectable concentration were considered to be at the minimum detectable concentration for averaging purposes Averages calculated using results below the minimum detectable concentration level were identified with a less than (<) sign ChemRisk included the < sign in reporting their compilation of annual average results from the Environmental Monitoring Reports

Table 2 Reported Annual Beryllium Releases from the Rocky Flats Annual Environmental Reports Compiled in Table 3-3 of the Phase I Task 5 Report (ChemRisk 1994a)

Year	Reported Release (g)	Year	Reported Release (g)
1971	16	1981	0 2
1972	<2 0	1982	0 1
1973	<7 1	1983	0 1[a]
1974	<10	1984	0 3
1975	<5 2	1985	0 5
1976	<3 7	1986	0 1
1977	<4 9	1987	0 2
1978	<17	1988	0 1
1979	<1 5	1989	0 6
1980	<1 1	Total 1971-1989	70 5

a The Annual Environmental Monitoring Report estimated an annual emission total for 1983 as a negative number

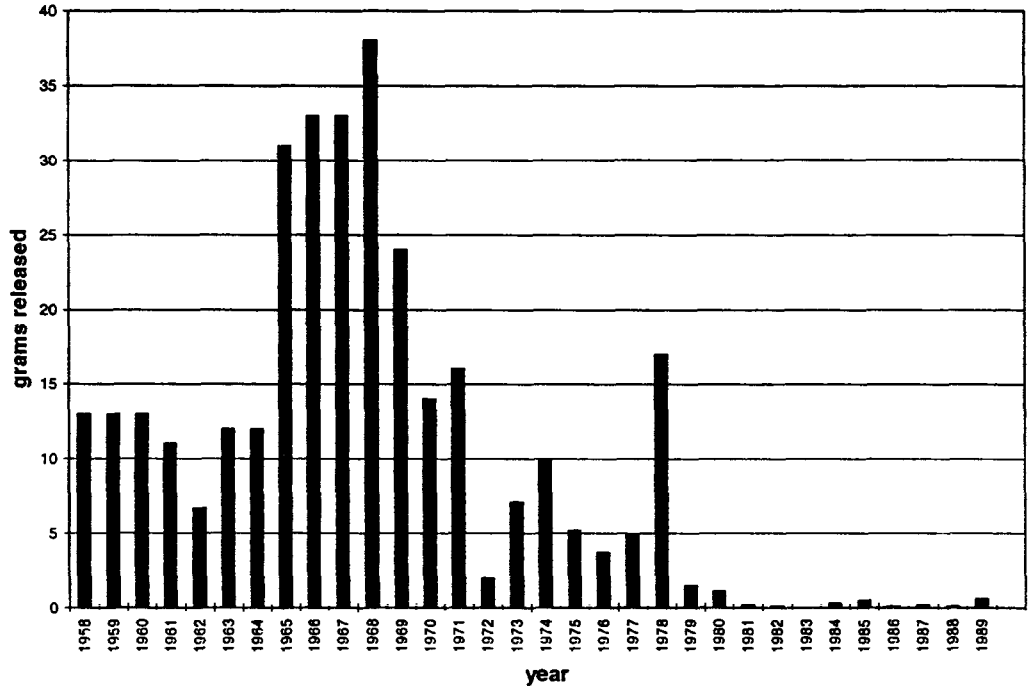

Figure 1 Annual release estimates for beryllium as estimated by ChemRisk (1994a)

The annual emission total for 1983 is a negative value of – 0 1 grams The explanation in the annual report suggested that the level in air sampled this year could not be distinguished from background levels (ChemRisk 1994a) Explanation for differences from year to year are not offered in the Phase I Task 5 report Documented changes in production and upgrades in cyclone separators and exhaust filtration are described but do not appear to correspond to the

release estimates (ChemRisk 1994a) Releases in the 1960s are probably related to peak production of beryllium parts Changes in exhaust ventilation were made in 1964, 1974, and several times in the 1980s (Holwager 1996) The variability in sample measurements was high largely because of averaging measurements of relatively low concentrations in large volumes of exhaust air Changes in analytical methods, ways of averaging effluent measurements, adjustments for background levels, and changes in methods that altered the degree to which beryllium solubilized in the samples are examples of reasons for the variability of the release estimates (Barrick 1997, Daugherty 1997) Quality assurance for effluent measurements are further discussed in the Phase II Task 4 Report (Rope et al 1997)

Uncertainty in the Source Term Estimates

Uncertainty associated with the beryllium source term estimates was characterized using the same approach applied to plutonium and uranium sample measurements and release estimates Uncertainties in exhaust and sample flow rate estimates, and in analytical results were combined and the total uncertainty was estimated using Monte Carlo methods (ChemRisk 1994a, Appendix G) The Task 5 report (ChemRisk 1994a) also contained a description of another source of uncertainty, as follows

> The beryllium release summaries suggest extremely low environmental emissions of beryllium averaging in the tens of grams or less annually A 1980 plant internal letter indicated that, based on an evaluation by the plant's General Service Laboratory, use of the minimum detectable amount (MDA) value for beryllium at each effluent measurement location would result in a calculated minimum beryllium discharge per month of 0 4 gram (Hornbacher 1980) This would lead to a reported yearly minimum discharge of about 4 to 5 grams even if none of the samples had a positive result. The information that was reviewed suggests that the beryllium data handling practices may have led to the reporting of annual emissions that were higher than the actual releases However, given the low magnitude of the reported emissions, the uncertainty introduced by this practice has not been characterized

ENVIRONMENTAL MONITORING FOR BERYLLIUM

Beryllium in Surface Water

Beryllium had the potential to be transferred offsite in surface water, and this pathway was investigated in Phase I Beryllium has been monitored in water effluent since 1980 (ChemRisk 1994a) Routine surface water monitoring for beryllium has always shown less than 0 05 mg per liter of water, which is the analytical detection limit

ChemRisk reported that available data on surface waterborne releases from the Rocky Flats Plant were not sufficient to develop direct estimates of release for beryllium They concluded that, "the only information available for addressing past releases from the plant would be measurements of beryllium in reservoir sediments" (ChemRisk 1994a) The beryllium compounds of concern are not very water soluble and would be expected to bind to sediments and soils Beryllium concentrations in the sediments of Great Western Reservoir

and Standley Lake are similar to background levels and concentrations found in soil and sediment samples from other Rocky Mountain regions (ChemRisk 1994a, EPA 1975, DOE 1995a)

Historically inhalation of beryllium has been a much greater human health concern than ingestion, in part because less than 1% of ingested beryllium is absorbed through the gastrointestinal tract (EPA 1991) Beryllium does not bioaccumulate in fish Releases of beryllium to surface water were not evaluated further because of a lack of source term and effluent and environmental monitoring data insufficient evidence of accumulation in soils and sediments, and the low solubility and gastrointestinal absorption of beryllium

Beryllium in Soil

Beryllium concentrations in soil are of interest because resuspension of beryllium in soil is of potential concern and a pattern of beryllium contamination in soil could reveal information about discharge from the plant Beryllium sources that might affect concentrations in soils at the Rocky Flats Plant include

- Operations at the plant
- A beryllium ore industry located 2 km east of the plant
- A beryllium ceramics industry 15 km south of the plant
- Coal burning and other combustion sources near the plant
- Beryllium in gravels brought into the site
- Naturally occurring beryllium (Barrick 1982, Kray 1992)

A study to characterize sources of beryllium and to quantify environmental beryllium contamination in soil near the RFP was conducted in 1981 (Barrick 1983) This study reported an estimate of 196 g for the total amount of beryllium exhausted from all buildings that processed beryllium during the 24-year period 1958 to 1982 This estimate included releases from two reported filter-fire accidents in February 1978, which released 14 5 g of beryllium from the main beryllium production building, Building 444 In this study 241 soil and rock samples from the site and from nearby areas were obtained Deeper samples were taken at 5 to 10 cm to establish geological background levels of beryllium The study concluded that RFP-originated beryllium could not be distinguished from geological, naturally occurring beryllium taken on lands outside plant property The survey found that natural gravels and an estimated 36 million kg of gravels brought in and added to Rocky Flats Plant surfaces have the highest and most variable beryllium concentrations The mean concentration in these gravels is 1 1 $\pm$ 1 4 $\mu g\ g^{-1}$ of soil (parts per million or milligrams per kilogram of soil) The background beryllium concentrations in soil (Rocky Flats alluvium) averaged 0 64 $\pm$ 0 07 $\mu g\ g^{-1}$

In what appears to be an earlier draft of these results Barrick (1982) suggested that atmospheric transport of beryllium to soils surrounding the plant had not occurred because no surficial soils near the plant were found to have elevated beryllium concentrations The mean level in soils in the plant area was reported to be 0 6 $\mu g\ g^{-1}$ and ranged from 0 2 to 1 1 $\mu g\ g^{-1}$ Higher levels found near roads and buildings were attributed to surficial gravel aggregates which had the highest background or natural beryllium levels

One accumulation of beryllium in soil that likely originated from Rocky Flats Plant operations was found 30 m from the stack of a plant that processed beryllium The samples

at this location were 44 to 69 μg g^{-1} of soil above background. Subsequent to this study, more samples were taken at various depths to try to determine when the accumulation in soil had occurred A high beryllium soil concentration was found in a 10 by 10 m^2, adjacent to door 10 of Building 444 The beryllium contamination was found within the top 5 cm of soil and ranged from 1 to 114 μg g^{-1} The pattern of contamination suggested the source of the beryllium release was door 10 and not the nearby stack for Building 444 or the filter plenum room The study's authors recommended removal of 1 m^3 of contaminated soil (Barnick 1983) Before 1970, chlorinated hydrocarbon solvent that had been used to rinse beryllium parts was disposed of by pouring it on soil outside door 10 on the south side of Building 444. A special study to sample beryllium in air near this solvent disposal site was done in the summer of 1977 Filters were collected weekly from an air monitor mounted 3 feet above ground level The detection limit was reported to be approximately 7 5× 10^{-9} μg m^{-3} The maximum level reported at this location was 2 3 × 10^{-3} μg m^{-3} and the average was 9 × 10^{-4} μg m^{-3} Because the air concentrations averaged about 9% of the ambient air standard of 0 01μg m^{-3}, soil removal was not recommended at that time (Barker 1978)

The Colorado Department of Public Health and Environment (CDPHE) conducted studies on beryllium in soil in 1971 and 1989 In 1992, at the request of Bob Quillan, (a Health Advisory Panel member representing the CDPHE), a discrepancy in the 1971 and 1989 beryllium soil sampling results were evaluated by CDPHE personnel (Quillan 1992) The study done in 1989 reported 21 results, all less than the analytical detection limit of 2 7 μg g^{-1} The 1971 data consisted of 13 results, ranging from 2.0 to 60 μg g^{-1} with no analytical detection limit reported The pattern of positive values seen in 1971 was not consistent with what would be expected if the beryllium in the soils had been deposited because of atmospheric dispersion from the RFP Spatial variations did not indicate a plume of beryllium from the plant operations. The 1989 dataset was judged to be more credible because of better documentation of analytical procedures, more rigorous quality assurance and improved analytical methods and equipment (Kray 1992)

To investigate potential contamination of surface soils from windborne dispersal in Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) Operable Unit (OU) 3 (offsite), the distribution of metals in OU 2 (onsite areas) was evaluated If contamination of soils onsite was due to activities at the plant, then the soil sampling results were expected to show a distinct spatial distribution trend of decreasing concentrations with increasing distances from areas where beryllium was used The CERCLA program personnel reasoned that if metal contamination of soil in onsite areas (OU 2) was at background concentrations or appeared to be a result of localized incidents of contamination, and if no spatial trends could be identified, then contamination in offsite (OU3) soils was unlikely and sampling of OU 3 soils (at distances further out) would not be warranted (DOE 1994) Samples were compared with results from two studies of background concentrations the Rock Creek and the Background Soils Characterization Project The study found a mean beryllium concentration in OU2 soils of 0 68 μg g^{-1} with a standard deviation of 0 21 μg g^{-1} and a coefficient of variation of 0 31 One extreme value of 1 50 μg g^{-1} was determined to be an outlier The beryllium concentrations in OU 2 soils were similar to those for Rock Creek soil samples which had a mean value of 0 68 μg g^{-1} and a maximum concentration of 0 96 μg g^{-1} The Background Soils Characterization Project study showed a

similar mean of 0 66 μg g ^{1} The U S Geological Survey (USGS) geometric mean for beryllium concentrations in the Front Range soil was 1 20 μg g^{-1} Beryllium concentrations in the OU2 soil did not appear to be above background No spatial trends in the data, or recognizable plume were apparent and no preferred direction of beryllium in soils was observed This study did not provide evidence of airborne contamination (DOE 1995a, Allen and Litaor 1995)

The results of soil monitoring conducted as a part of investigations of CERCLA OU 5 (Woman Creek Drainage) and OU 6 (Walnut Creek Drainage) do not suggest a windborne deposition pattern (DOE 1995b,1996)

Samples of cottonwood leaves, collected from trees growing on soils with 0 1 – 1 0 μg g^{-1} beryllium contained beryllium concentrations that were only slightly correlated (r = 0 25) with the concentration in the soil This observation led researchers to decide that leaf surveys would not be useful as indicators of soil contamination (Barrick 1983), although analysis of cottonwood trees has been used to locate beryllium ore deposits in the USSR

Taken together the soil data suggest that beryllium deposited on soil from RFP releases is not distinguishable from beryllium in the soil from natural and other sources

Beryllium in Ambient Air

Historical ambient air monitoring for beryllium in the vicinity of the Rocky Flats Plant is reviewed and put into perspective in the Phase II Task 4 Report Evaluation of Historical Monitoring Data (Rope et al 1997) The beryllium ambient air data were not considered as a part of Phase I

The Dow Chemical Company site survey monthly reports from the 1950s contain some qualitative statements and a few quantitative measurements of beryllium in ambient air Routine monitoring was conducted from 1970 to 1976 and reported in the Dow Chemical Company Monthly Environmental Reports The Rocky Flats Plant beryllium releases were less than the EPA s discharge limit of 10 grams per stationary source for a 24-hour period (EPA 1973) in the 1970s It is likely that the ambient air monitoring results were not reported in the annual environmental reports because the results were thought to be low and the site was in compliance with EPA standards (Rope et al 1997)

Time trend analysis suggests that the concentrations in onsite air appear to be unrelated to the amount of beryllium released from the plant Resuspension of beryllium released to the soil from plant operations did not appear to contribute significantly to offsite air concentrations The monitoring data support the prediction that offsite air concentrations of beryllium are well below background concentrations (Rope et al 1997)

Beryllium in Waste

Beryllium was also present in waste some of which was discharged into the solar evaporation ponds ChemRisk described the disposal of waste from the Coors Porcelain Company at the RFP (ChemRisk 1992) Resuspension or leaching of beryllium in waste has not occurred in the past to an extent to warrant inclusion in this study

THE HEALTH HAZARD OF BERYLLIUM

This section includes a description of the regulatory standards for beryllium in air, evidence of carcinogenicity, and a review of the literature on chronic beryllium disease

Regulatory Guidelines for Beryllium

Because of beryllium's use in the nuclear weapons industry, the Atomic Energy Commission recommended occupational and community ambient air standards for beryllium in 1949 (Eisenbud et al 1949) that greatly reduced beryllium exposures in and around beryllium plants The community air standard became the first ambient air quality standard in the United States It preceded all others by about 25 years, and the standard remains unchanged to this day (Lang 1994) The ambient air standard, also called the 'neighborhood' air standard, limits beryllium concentrations in air surrounding factories to 0 01 $\mu g/m^3$, averaged over a 30 day period (EPA 1987a) The occupational limit is 0 002 mg/m^3 (Meyer 1994)

Beryllium Carcinogenicity

Numerous studies have shown that beryllium compounds are carcinogenic in experimental animals by several routes of exposure, including inhalation, however, there has been considerable debate as to whether beryllium can cause cancer in humans

A number of epidemiological studies have reported an increased risk of lung cancer in beryllium workers, but deficiencies in the studies have not allowed unequivocal conclusions to be made (Meyer 1994, IARC 1980, EPA 1987b) Criticisms include little or no consideration of smoking history, exposure to other potential lung carcinogens, and underestimating expected cancer deaths in control populations (Smith 1981, Meyer 1994)

In a review of the U.S Beryllium Case Registry Data, Hardy et al (1980) reported there was no evidence to support beryllium as a human carcinogen, but the authors recommended workers be studied

Four epidemiological studies conducted before 1970 did not clearly demonstrate a relationship between exposure to beryllium compounds and the occurrence of human cancer, but excess risk is suggested by the results of all of the studies (Wagoner 1980, Mancuso 1980, IARC 1980, EPA 1996)

Additional studies in the 1990s found excess risk of lung cancer in workers enrolled in the Beryllium Case Registry (Steenland and Ward 1991) Occupational exposure to beryllium compounds was said to be the most plausible explanation for the increased risk of lung cancer observed in these studies (Ward et al. 1992)

Four International Agency for Research on Cancer (IARC) working groups (in 1972, 1980, 1987, and 1993) reviewed the animal and epidemiological data on beryllium carcinogenicity The first working group considered the epidemiological studies available at that time as inadequate to evaluate the human carcinogenicity In 1980 and 1987 the working group concluded that beryllium was carcinogenic to animals but that epidemiological evidence was limited They classified beryllium as a suspect human carcinogen. Epidemiological evidence was again carefully scrutinized by the IARC working group convened in 1993 The proceedings of the 1993 conference, in IARC Monograph Volume 58, states that compounds of beryllium are carcinogenic in animals by a number of routes, and several beryllium

compounds produce lung tumors in rats exposed by inhalation The working group concluded that there was sufficient evidence in experimental animals for the carcinogenicity of beryllium and beryllium compounds After a review of all available epidemiological studies, the working group concluded that there was also sufficient evidence in humans for the carcinogenicity of beryllium and beryllium compounds However, controversy about the classification of beryllium as a human carcinogen continues

Studies implicating beryllium as an occupational carcinogen have examined lung cancer in cohorts exposed in the 1930s and 1940s before industrial hygiene controls were in place, when concentrations were orders of magnitude higher than permitted today Statistically significant increases have been difficult to demonstrate in workers exposed to lower levels (Jameson 1996)

Currently, beryllium is classified by the EPA as a B2 probable human carcinogen, primarily on the basis of sufficient evidence from animal experiments Evidence in humans is considered inadequate or limited (EPA 1996) Beryllium risk values are scheduled to be reevaluated by the EPA in 1997

Chronic Beryllium Disease

The potential for historical releases of beryllium from the RFP to have caused chronic beryllium disease in offsite individuals was not addressed in Phase I

Chronic beryllium disease, also called berylliosis, is a progressive granulomatous disease Although the lung is primarily involved, it is a systemic disease and granulomatous inflammation may involve other organs A delayed-type hypersensitivity reaction is thought to play a central role in the pathogenesis of chronic beryllium disease Sensitization to beryllium can be detected by measuring in vitro proliferative responses of bronchioalveolar lavage lymphocytes or peripheral blood lymphocytes to beryllium Clinical and experimental animal data on chronic beryllium disease support an immunologic, hypersensitivity mechanism for chromic beryllium disease Information consistent with such a mechanism includes (a) the insidious nature of the disease, (b) a long latency between exposure and onset, (c) the granulomatous nature of the lung lesions that develop, (d) berylliosis patients show delayed skin hypersensitivity reactions to beryllium compounds (e) peripheral blood lymphocytes and bronchoalveolar lymphocytes in people with chronic beryllium disease undergo blast transformation and release of migration inhibition factor after exposure to beryllium in vitro, and (f) the lack of a dose response relationship (Deodhar and Barna 1991, Hardy 1980, Kriess et al 1993a, Aller 1990 Clarke et al 1993)

Susceptibility to sensitization is likely to have a genetic basis Recently, a gene was identified in people with sensitivity to bervllium (Richeldi et al 1993) It was concluded that people with this gene have a significantly increased probability of developing sensitization than those without it (Newman 1993) However it appears that about 30% of the population has the gene and at most only about 10–15% of exposed workers become sensitized (Lang 1994)

Most commonly researchers estimate that 1 to 5% of beryllium-exposed workers develop chronic beryllium disease (Eisenbud and Lisson 1983 Meyer 1994, EG&G Rocky Flats 1991) Sensitization rates may be higher Kriess et al (1993b) reported 2 9 to 15 8% for beryllium exposed persons

Most cases of chronic beryllium disease have occurred in people working in industries processing or using beryllium, however, cases of chronic beryllium disease have been reported in people living near processing plants and in families of beryllium workers, perhaps from exposure to airborne beryllium carried from a plant or from handling contaminated worker's clothing Chronic beryllium disease has also developed in peripheral workers, people in nonprocessing areas of factories, who were likely exposed to very small amounts of beryllium (Drury et al. 1978, Hardy 1980, Hasan and Kazemi 1974, Meyer 1994) Although Kriess et al (1993b) reported that the degree of beryllium exposure was associated with disease rates, they found that sensitization occurred in workers with exposures as short as a 1 month or in people with unrecognized exposure

The occurrence of beryllium disease in those with inadvertent or seemingly trivial exposure has been reported in secretaries and security guards at the RFP (Kriess et al 1993b) and other facilities (Lang 1994), a janitor in a ceramics company (Lang 1994), and in members of worker's households and neighbors around beryllium extraction plants (Eisenbud et al 1949, Eisenbud and Lisson 1983, EPA 1996) The latter are termed neighborhood cases, which are cases of chronic beryllium disease that occur in people living in the vicinity of the beryllium plants (EPA 1987b)

In a report summarizing the relationship between the incidence of nonoccupational related cases of chronic beryllium disease and the levels of atmospheric contamination, Eisenbud et al. (1949) observed that the distribution of the cases with respect to the plant indicated that the incidence of disease was a function of the concentration to which the residents were exposed The incidence of disease within one-quarter of a mile was about 1% or 5 of 500 people (Eisenbud et al. 1949) The cases of chronic beryllium disease in the 1930s and 1940s in Salem, Massachusetts, occurred almost entirely in fluorescent lamp manufacturing workers except for three neighborhood cases. a night watchman, a near neighbor, and a housewife with two young women living in her home who were fluorescent lamp workers. Protection was minimal and workers were exposed to high levels of beryllium phosphors (Hardy 1980)

Chronic beryllium disease occurred as an epidemic in the 1940s, leading to the establishment of the Beryllium Case Registry in 1951 The Case Registry is a file for cases of acute and chronic beryllium disease, now maintained by the National Institute of Occupational Safety and Health in Cincinnati (Lang 1994, EPA 1987b) In 1983, Eisenbud and Lisson reviewed the Beryllium Case Registry's 224 acute and 622 chronic cases These cases included 577 occupational and 65 chronic beryllium disease cases attributed to ambient air pollution, 42 were attributed to ambient air exposure in areas in the vicinity of beryllium plants and 23 to exposure to dust brought home on workclothes They reported no new cases for individuals exposed after 1972 and believed that control measures implemented in the 1950s had reduced chronic beryllium disease despite a marked increase in the use of beryllium However, results of more recent research and clinical work have led to questions about the effectiveness of beryllium control measures and standards on reducing the incidence of chronic beryllium disease

Although many researchers have praised the apparent effectiveness of the air standards for beryllium and have asserted that no new cases of beryllium disease have occurred since observance of these limits (Hurlbut 1974), others believe that the occupational standards

may not be protective for sensitization (Newman 1993, Lang 1994) and the limit designed to protect the general public may not be low enough (Clarke et al 1993) The EPA considers the ambient air standard protective for the public with ample margin of safety (EPA 1987b)

Lee Newman, of the Occupational and Environmental Medicine Division of the National Jewish Center for Immunology and Respiratory Medicine in Denver, was reported to believe that with increasing use of beryllium in industry, the absolute number of cases can be expected to increase He said, "even with careful ventilatory controls and monitoring, cases of chronic beryllium disease will continue to occur such hypersensitivity can develop in some individuals following even low-level exposures, well within permissible exposure limits" (Lang 1994) Evidence exists for biological responses and possible sensitization occurring after exposure to levels far below the current threshold limit values (Clarke et al 1993)

No dose response relationship (dose is usually measured by duration of exposure) has been established for this chronic beryllium disease, which is interpreted as involving a delayed-type hypersensitivity so that even very low exposures may be sufficient to induce it Chronic beryllium disease can develop in people with relatively low exposures, whereas nonsensitized people experiencing high exposures may not develop the disease (Deodhar and Barna 1991, Wagoner et al 1980, Mancuso 1980) Even slightly exposed individuals, such as neighborhood cases sometimes show severe clinical forms of the disease (Hardy 1980, Eisenbud et al 1949)

Recent studies published in Kriess et al (1993b) suggest that both individual sensitivity and degree of exposure or exposure circumstances are important in determining the risk of developing chronic beryllium disease Although chronic beryllium disease cases have been associated with trivial or unrecognized beryllium exposure, chronic beryllium disease rates were higher in workers with presumed greater beryllium exposure, seeming to challenge the immunological dogma of no dose-response in chronic beryllium disease

In general, the most appropriate end point for risk assessment is the effect that occurs at the lowest exposure Because chronic beryllium disease can develop with very low-level exposure it may be a better end point than lung cancer for assessing risk to low-level exposures However, chronic beryllium disease may not be dose-related and the percentage of an exposed population that might be expected to develop the disease at a given exposure level is not known (Jameson 1996) Conducting a quantitative risk assessment is not feasible because of the lack of dose-response

In light of the complexity and apparent immunological (no dose-response) nature of chronic beryllium disease the Health Advisory Panel for the Rocky Flats Health Studies chose lung cancer rather than chronic beryllium disease, as an end point for risk assessment

PHASE I EXPOSURE AND RISK CALCULATIONS

For Phase I beryllium releases and transport in air were modeled using assumptions that are described in detail in the Task 6 Report (ChemRisk 1994b) The predicted annual air concentrations were presented as concentration isopleths on maps of the site and surrounding area In calculating deposition of airborne contaminants it was assumed that the particles released were submicron in size because of HEPA filtration Small depositional velocities were used and dry and wet deposition were considered The calculations and uncertainties are described in Section 3 of the Task 6 Report (ChemRisk 1994b) Nine

potentially important exposure pathways for beryllium were identified and listed in the Phase I Task 6 report Using the predicted air and soil concentrations for Sector 12 (southeast of RFP), the exposure equations provided in Appendix I, and parameters in Appendix J, pathway specific doses were estimated in milligrams per kilogram per day using Monte Carlo simulation The exposed individual was assumed to be an adult consuming a typical amount of air, food, water, vegetables, and soil. Assumptions about time spent outdoors, consumption of local produce, beef, and milk, and exposure are described in the Task 8 report (ChemRisk 1994c)

Doses of beryllium in units of microgram per year were calculated and reported in the Task 8 Report (ChemRisk 1994c) The highest (geometric mean) dose reported in Appendix L for Sector 12 for 1968 was $1\ 1 \times 10^{-4}$ μg y^{-1} for inhalation and $9\ 7 \times 10^{-6}$ μg y^{-1} for all ingestion pathways combined The resulting risk for 1 year of exposure reported in Appendix N was (geometric mean) 1×10^{-12} (GSD = 2 6) for inhalation risk and 2×10^{-12} (GSD = 2.8) for ingestion risk

PHASE II EXPOSURE AND RISK CALCULATIONS

Annual release estimates, release points, and the percentage contribution to the total releases from the site, reported in Phase I (ChemRisk 1992, ChemRisk 1994a) were used for Phase II calculations The greatest release occurred in the year 1968 The greatest source of beryllium was operations in Building 444 The annual beryllium emission estimates for 1960-1970 were calculated from data compiled from sample data logbooks and using exhaust volume estimates made in ChemRisk (1992). Because emissions data were lacking before 1960, estimates for 1958 and 1959 were made from those calculated for 1960 Estimates for 1971–1989 were taken from the RFP Annual Environmental Monitoring Reports ChemRisk independently calculated releases for 1 year during this period (1984) and found good agreement between their calculation and the value reported in the 1984 Annual Environmental Monitoring Report The source term estimates consider releases during fires that have occurred in Building 444

For this assessment, inhalation of air is the exposure pathway of concern Beryllium is not well absorbed after ingestion Beryllium is relatively immobile in surface water, tending to absorb to soils and sediments, and would not be predicted to transport offsite to a great extent Beryllium intake from ingestion of vegetation subject to deposition from the air, livestock inhaling air, surface water, soil and sediment, livestock ingesting soil or sediment, vegetation grown on soil, and livestock ingesting vegetation grown on soil could be evaluated but would be expected to contribute only a small amount to overall risk.

In Phase I, a suggestion was made to include the exposure route of dermal contact and wound entry The response ChemRisk gave to this suggestion in Appendix O of the Task 8 Report (ChemRisk 1994c), which asserts that for the offsite population at large, dermal absorption and wound entry are not likely, is appropriate Although these routes are important for occupational exposure, absorption through the skin or through wounds would be an extremely rare occurrence for individuals offsite

Cancer Potency Determination

The EPA weight-of-evidence classification for beryllium is B2, a probable human carcinogen B2 carcinogens are defined by the EPA as chemicals with sufficient evidence of carcinogenicity in animals with inadequate or a lack of human data

The estimate of the excess lifetime cancer risk is the product of the dose and the carcinogenic potency slope factor (SF)

Excess cancer risk = beryllium exposure concentrations × slope factor

Cancer slope factors are usually estimated from animal studies using mathematical models, most commonly the linearized multistage model, for estimating the largest possible linear slope (within the 95% confidence limit) at extrapolated low doses that are consistent with the data The SF is expressed in units of the inverse of milligram intake per kilogram body weight per day (kg d mg^{-1}) It represents the 95% upper confidence limit of the probability of a carcinogenic response per daily unit intake of a chemical over 70 years The SF (and risk) is characterized as an upper-bound estimate The true risk to humans, while not identifiable, is not likely to exceed the upper bound estimate

The inhalation unit risk factor is the risk per unit concentration in air, calculated by dividing the SF by 70 kg and multiplying by the inhalation rate ($20 m^3$ d^{1}) (EPA 1995)

$$UR = \frac{SF\ BR}{BW\ CF} \quad (1)$$

where

UR = unit risk (m^3 μg^{-1})
SF = slope factor (kg d mg^{-1})
BR = breathing rate (m^3 d^{1})
CF = correction from mg to µg (1×10^{-3})

Using this relationship, a SF of 8 4 mg/kg/day was calculated from the mean of the unit risk values published in Integrated Risk Information System (IRIS) (EPA 1996)

For the quantitative estimate of the carcinogenic risk from inhalation exposure, the inhalation unit risk value was calculated using human occupational epidemiological data of Wagoner et al (1980) Justification for this approach included the fact that humans are most likely to be exposed by inhalation to beryllium oxide rather than other beryllium salts, and animal studies of beryllium oxide have involved intratracheal instillation rather than inhalation

Relative risk estimates were derived from the smoking adjusted lung cancer death data The relative risk estimates ranged from 1 36 to 1 44, and the 95% confidence limits of these estimates, 1 98 and 2 09, were used to estimate the lifetime cancer risk The estimates were based on one dataset using a range of estimated exposures and exposure durations The effective dose was calculated by adjusting for durations of daily (8/24 hours) and annual (240/365 days) exposure and the fraction of the lifetime at risk (duration of employment) Because of uncertainties in the bervllium exposure levels and exposure times unit risks were derived using two estimates each of concentration fraction of lifetime exposed and relative risk These data are summarized in Table 3 The recommended value for use in risk

assessment published in IRIS is 2.4×10^{-3}, the arithmetic mean of the eight derived unit risks The values are conservative, calculated using the 95% confidence limit of the relative risk estimates Absorption of beryllium is taken into account in developing unit risk levels Although based on human data, which generally provide for more confidence than animal data, the quality of the study on which the estimates are based is considered poor because the study was confounded by several variables A quantitative assessment based on animal studies was reported to have resulted in a similar estimate of risk (EPA 1996, EPA 1987b)

Table 3 Values from Human Inhalation, Occupational Exposure Studies Used to Calculate Unit Risk Values (EPA 1987b; EPA 1996)

Workplace beryllium concentration ($m^3\ \mu g^{-1}$)	Fraction of the lifetime	Dose ($m^3\ \mu g^{-1}$)	95% upperbound estimate of relative risk	Unit risk ($m^3\ \mu g^{-1}$)
100	1 0	21 92	1 98	1.61×10^{-3}
100	1 0	21 92	2 09	1.79×10^{-3}
100	0.25	5 48	1 98	6.44×10^{-3}
100	0.25	5 48	2 09	7.16×10^{-3}
1000	1 0	219 18	1.98	1.61×10^{-4}
1000	1 0	219 18	2 09	1.79×10^{-4}
1000	0 25	54 79	1 98	6.44×10^{-4}
1000	0 25	54 79	2 09	7.16×10^{-4}

The *Health Assessment Document for Beryllium* (EPA 1987b) describes deficiencies of the epidemiological data, efforts by the EPA's Carcinogen Assessment Group to adjust the data for use in calculating cancer potency, and assumptions and models used to extrapolate from high occupational exposures to low-level exposures

According to the information in IRIS, an EPA workgroup last assessed the beryllium risk values in 1988 The EPA is currently reevaluating beryllium cancer risk as a part of the IRIS Pilot Project The reevaluation is not undergoing internal peer review A preliminary draft may be available to the public in June 1997 (Bayliss 1997, Bruce 1997)

Uncertainties in the Slope Factors

Slope factors are uncertain The values used for this assessment are those recommended by the EPA in the IRIS Database (EPA 1996) They were derived from a range of epidemiological data, which is summarized in Table 3 There are obvious limitations to developing values from the results of a single worker epidemiological study with confounding factors and limitations of its own Uncertainties associated with the concentrations of beryllium in the workplace, duration of exposure, dosimetry, and other assumptions used in determining the unit risk values are discussed in EPA (1987b) but were not quantified

The relative risk estimates were used to provide a probable range and central value rather than just a 95% confidence limit value In the occupational epidemiological study on which the cancer potency determination was based, a range for median exposure of 100 to 1000 $\mu g\ m^{-3}$ was determined Furthermore, an assumption was made that the ratio of exposure duration to years at risk ranged from 0.25 to 1 0 The mean of the potency factors

derived using these assumptions was reported in IRIS (EPA 1996) The maximum and minimum values (EPA 1987b) can be used to calculate a minimum and maximum SF The maximum risk per microgram per cubic meter value of $7\ 16 \times 10^{3}$ corresponds to a SF of 25 (kg d mg^{-1}) and the minimum risk per microgram per cubic meter value of $1\ 61 \times 10\text{-}4$ corresponds to a slope factor of 0 56 (kg d mg^{-1}) These values were used to approximate an uncertainty distribution for the SF assuming a triangular distribution with the most likely value being 8 4 kg d mg^{-1}

ENVIRONMENTAL TRANSPORT MODELING

Atmospheric releases of beryllium during routine operations at the RFP primarily occurred from two release points roof vents on Building 776 and Building 444 Other minor release points were also identified in Phase I reports In this section, we describe our approach to estimating atmospheric dispersion of beryllium for the years 1953–1988 and the uncertainty associated with concentration estimates in the model domain We have incorporated estimates of the quantities of beryllium released to the atmosphere provided by ChemRisk in Phase I and summarized in a previous section We use these dispersion estimates, along with the exposure scenarios developed and carcinogenic SFs, to calculate risk for selected receptors in the model domain

Atmospheric Model Selection

Five atmospheric transport models considered for use in this study were evaluated in Rood (1996) (1) the Terrain-Responsive Atmospheric Code (TRAC) (Hodgin 1991), (2) the Industrial Source Complex Short Term Version 2 (ISC) (EPA 1992) (3) Regional Atmospheric Transport Code for Hanford Emission Tracking (RATCHET) (Ramsdell et al 1994), (4) TRIAD (Hicks et al 1989), (5) and INPUFF2 (Petersen and Lavdas 1986) The purpose of the model comparison study was to determine what models if any, performed best in the Rocky Flats environs for a given set of modeling objectives These data along with other studies were used to established the uncertainty one might expect from a model prediction

Model evaluations were based on how well predictions compared with measured tracer concentrations taken during the Winter Validation Tracer Study (Brown 1991) conducted in February 1991 at the RFP The study consisted of 12 separate tests, 6 were conducted during nighttime hours 4 during daytime hours and 2 during day-night transition hours For each test, an inert tracer (sulfur hexafluoride) was released at the RFP at a constant rate for 11 hours Two sampling arcs 8 and 16 km from the release point, measured tracer concentrations every hour for the last 9 hours of each test period Seventy-two samplers were located on the 8-km arc, and 68 samplers were located on the 16-km arc Predicted concentrations were then compared to the observed tracer concentrations at each of the samplers

Modeling objectives for the comparison study were based on the premise that identifying locations of individual receptors on an hour-by-hour basis was unlikely Instead it was more likely to identify receptors (hypothetical or real) who were present at a fixed location for the duration of a release event The minimum time scale of historical release events at RFP ranged from one to several days Release events modeled for the Winter Validation Tracer Study were 9 hours in duration If we assume the receptor is fixed for a time period of at

least 9-hours, then the time-averaged concentration (9-hour average) is an appropriate modeling objective rather that comparing hourly average concentrations Therefore, models were evaluated based on their performance in predicting time-averaged concentrations at fixed sampler locations in the model domain (9-hour average concentration at each sampler paired with the corresponding predicted value) We also considered the arc-integrated concentration The arc-integrated concentration was the 9-hour average ground-level concentration integrated across the 8 and 16-km sampling arc The latter performance objective provides a measure of the vertical dispersion component of the models and the ground-level tracer mass, 8 and 16-km from the release point. Data sets for the time-averaged concentration were limited to only those points where the predicted (C_p) and (C_o) observed concentration pair were greater than the time-averaged minimum detectable concentration.

Fifty percent of the time-averaged model predictions were within a factor of 4 of the observations Ratios of the predicted-to-observed ratio (C_p/C_o) ranged from 0 001 to 100 and tended to be higher at the 16-km arc than the 8-km arc except for RATCHET, which generated consistent results at both sampling arcs Geometric mean C_p/C_o ratios ranged from 0 64 (TRAC) to 1 5 (ISC), and geometric standard deviations ranged 4 7 (RATCHET) to 6 5 (ISC) The RATCHET model had the highest correlation coefficient for the 8-km (0 59) and 16-km (0 54) sampling arc followed by TRIAD and INPUFF2

Arc-integrated results (Figure 2) showed INPUFF and TRIAD had the highest correlation coefficients, but correlation coefficients were not significantly different from the other models Qualitatively, the predictions made by the RATCHET model appear to best match the observations The slope of the regression line was closest to that of the perfect correlation line (solid line in Figure 2) The ISC model tended to overpredict arc-integrated concentration, and the TRAC model showed the greatest variability

The results reported in Rood (1996) indicated no one model clearly outperformed the others However, the RATCHET, TRIAD, and INPUFF2 models generally had lower variability (indicated by lower geometric standard deviations of C_p/C_o ratios) and higher correlation coefficients compared to those of ISC and TRAC models It is desirable in a study such as this to choose a model that has the least amount of variability when comparing model predictions to observations In addition, the model selected should have a level of complexity that is consistent with available data. The TRAC model is the most complex in terms of its treatment of the atmospheric dispersion process in complex terrain, but the study showed model performance was no better than the other models In addition, the availability of meteorological data needed to fully use the capabilities of this model are lacking The straight-line Gaussian plume model, ISC tended to overpredict concentrations and was also limited to only one meteorological recording station in the model domain Available meteorological data for this study period may include two meteorological recording stations, one at the RFP and the other at Denver Stapleton International Airport Therefore, a model that may include multiple meteorological recording stations in the model domain is desirable The use of multiple meteorological recording stations will allow for spatially varying wind field in the model domain.

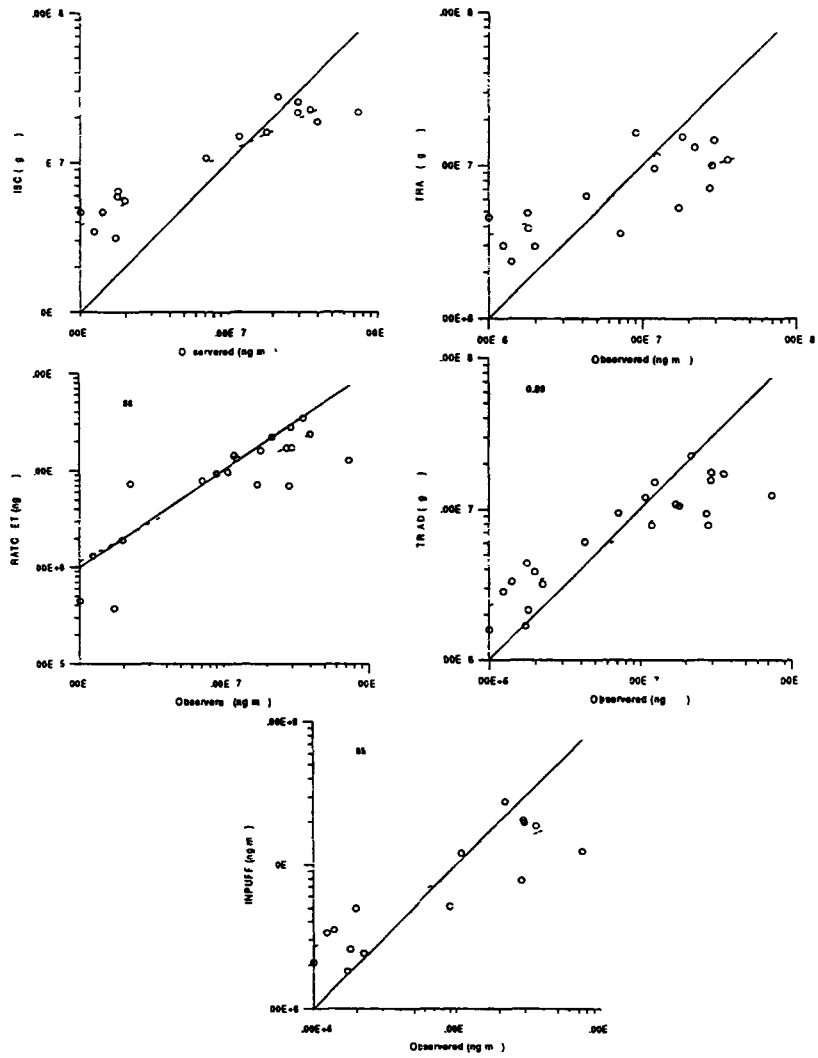

Figure 2 Observed arc integrated concentration as a function of predicted values for the five models compared using the Winter Validation Data Set Correlation coefficients were for the log transformed data The solid line represents perfect correlation between predicted and observed values The dashed line represents the log transformed regression fit

The three models RATCHET INPUFF2 and TRIAD performed comparably and were considered viable candidates for atmospheric dispersion estimates We chose the RATCHET code for modeling routine releases of beryllium for the following reasons

- The model was easily configured for long-term (annual average) dispersion estimates
- Spatial differences within the model domain are accounted for (i e surface roughness meteorology)
- Algorithms to compute plume depletion and deposition for fine particles are included (deposition must be computed outside the TRIAD and INPUFF2 codes)
- The model requires meteorological data in 1 hour increments which are the same as those given for typical airport observations

Corrections for model bias were made in the uncertainty analysis Features of the RATCHET model are summarized in Table 4

Table 4 Features of the RATCHET Model

Feature	Representation in RATCHET
Domain area[a]	2,100 km^2
Node spacing[a]	2,000 m
Source term	Hourly release rates
Meteorological data	Hourly
Surface roughness	Spatially varying
Wind fields	$1/r^2$ interpolation
Topographical effects	None explicit[b]
Wind profile	Diabatic
Stability	Spatially varying based on wind, cloud cover, and time of day
Precipitation	Spatially varying, three precipitation regimes with different precipitation rate distributions
Mixing layer	Spatially varying, based on calculated values for each meteorological station
Plume rise	Briggs' equation
Diffusion coefficients	Based on travel time and turbulence levels
Dry deposition	Calculated using resistance model
Wet deposition	Reversible scavenging of gases, irreversible washout of particles
Model time step	15 minute maximum, 15 second minimum
Output frequency[c]	Daily
Uncertainty	Options available for Monte Carlo simulation within the code

a Modified from the original RATCHET specification for use at Rocky Flats
b Terrain differences are not a model input. However, topographical influence on the wind field may be accounted for by incorporating multiple meteorological stations in the model domain
c Modified to output annual average concentrations at user specified grid nodes

Model Domain and Receptor Grid

The model domain (Figure 3) encompasses a 2,200 km^2 area (50 km north-south by 44 km east-west) The domain extends 28 km south, 12 km west, 22 km north, and 32 km east from the RFP Most of the Denver metropolitan area and the city of Boulder are included in the domain The domain was limited in its western extent because few receptors are present there and most of the contaminant plumes traveled east and southeast of the plant.

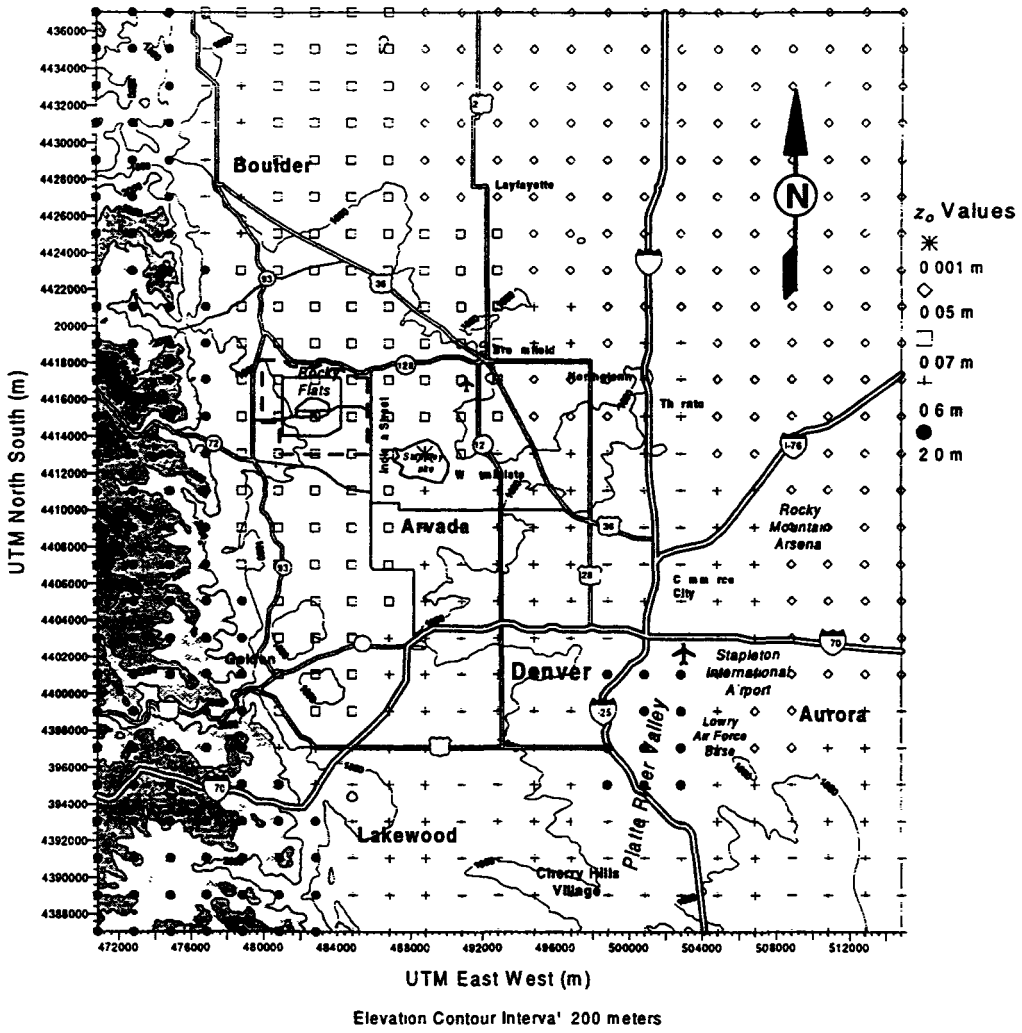

Figure 3 RATCHET environmental modeling grid and roughness length values (z_o) Symbols represent grid nodes and the z_o value assigned to the node

RATCHET uses two modeling grids Hourly meteorological records are used to estimate wind speed and direction stability and precipitation on the environmental grid in addition to surface roughness features The concentration grid has spacing one-half that of the environmental grid Ground-level concentrations and deposition are output at each of these grid nodes The environmental grid was set at 23 nodes east-west and 26 nodes north south with a grid spacing of 2,000 m The concentration grid has 45 nodes east west and 51 nodes north-south with a spacing of 1,000 m The southwest corner of the model domain has the UTM coordinates 470850 E and 4387050 N Release points are defined by distances (in kilometers) from a reference node The reference node for the environmental grid was (7,15) and (13,29) for the concentration grid and both have the UTM coordinates of 482850 E and 4415050 N

Figure 3 was generated using United States Geological Survey (USGS) 7 5-minute digital elevation models Topographic contours were based on an elevation grid spacing of 100 m Major roadways were digitized from USGS 1 100,000 digital line graphs Roughness lengths were defined for each environmental grid node and were separated into regions delineated by the dotted lines on Figure 3

Meteorology

Meteorological data for the operational period of Rocky Flats (1952–1988) are sporadic, incomplete, and of questionable integrity Requests for meteorological data from the RFP were initially made by ChemRisk during Phase I of the project ChemRisk was able to locate two letters from Dow Chemical to Dr Roy Cleare, Executive Director of the Colorado Department of Health, dated March 20, 1970, that contained wind speed and direction for varying time increments during the 1957 and 1969 fire incidents. Computer diskettes containing wind speed, wind direction, and precipitation measurements from October 1968 to May 1969 were also obtained These data were hourly observations taken approximately 15 minutes before the top of the hour and do not represent hourly average readings Although these data appeared to be climatologically reasonable, no records of instrument calibration or audits of the information were found Parameter resolution was very coarse (for example, wind direction resolution was 45 degrees) Five years (1987–1991) of high quality meteorological data taken at the 61-m tower at RFP were obtained and used by ChemRisk in Phase I of this project for predicting annual average concentrations from routine releases

An extensive data search was initiated in 1994 by *Radiological Assessments Corporation (RAC)* researchers to locate missing data and interview personnel who were involved with measurements at the site No new data were recovered, but several personnel reported problems with the recording instrumentation at the RFP, such as the measured wind direction being off by 180 degrees. Other data recorded from nearby Jefferson County Airport (about 8 km east of the plant) were obtained for the years 1968–1971 These data were only reported for the hours while the airport was open (06 00–23 00 local standard time) and were instantaneous measurements and not hourly averages as was typical of all airport data before the Automatic Surface Observation Site (ASOS) system was installed at most major airports In 1994, the RFP hired a subcontractor to compile, screen, validate, and analyze historical climatological data (DOE 1995c) A draft report was issued in February, 1995, the report contained monthly and annual summaries of wind speeds, wind directions, precipitation, temperature, and other parameters for the years 1953–1993 While these data are of interest and may be important for some aspects of modeling, they lacked the resolution required for detailed atmospheric transport modeling

We concluded that meteorological data taken during the time the RFP was operating were incomplete, unreliable, and unsuitable for atmospheric transport modeling during the period, 1952–1988 However, surrogate data spanning a different time period can be used to make annual average dispersion estimates for past releases We used this approach in our modeling effort

For our modeling effort, we used meteorological data spanning a 5-year period (1989–1993) taken at two recording stations located at the RFP and Denver Stapleton International Airport The EPA considers a 5-year database adequate for predicting annual-average air

quality impacts at a site Meteorological data from RFP were taken at the 10-m level from the 61 m tower located on the south side of the plant complex at UTM coordinates 482064 E 4414963 N Data recorded at this station included wind speed, wind direction temperature and other parameters (heat flux and standard deviation of wind direction) that were not used in these simulations The Denver Stapleton meteorological station was located 24 km east and 14 km south from the center of the model domain (RFP) These data included hourly measurements of wind speed, wind direction cloud cover, and precipitation It is known that meteorological conditions in the Denver metropolitan area can differ significantly from those at Rocky Flats (DOE 1980) Therefore, it is unreasonable to use meteorological data from Denver alone for simulations involving releases from Rocky Flats In these simulations, initial plume trajectories are primarily influenced by the wind direction at Rocky Flats Only after plume elements are transported to the Denver metropolitan area are trajectories and dispersion influenced by meteorological conditions present there

Data Processing

Meteorological data from 1989–1993 were obtained in electronic format from the Rocky Flats meteorologist These data were measured at a height of 10 and 61-m from a 61-m tower located at RFP Only data from the 10 m level were used in the simulations Each record represented the average over a 15-minute recording period and included wind speed and direction, temperature, heat flux, and standard deviations of these parameters Processed data suitable for use in EPA's ISC code were also obtained for the same time frame These data included stability class estimated by the lateral turbulence and wind speed method (standard deviation of the horizontal wind direction fluctuations) as described in EPA (1987c) and mixing height estimates The mixing heights were derived from linear interpolation for each 15-minute period from the rawinsonde data furnished routinely every 12 hours by the National Weather Service for Stapleton International Airport These data were used as default mixing-layer depths in RATCHET Mixing-layer depths are calculated hourly within RATCHET at each active meteorological recording station using a methodology described by Zilitinkevich (1972) The calculated or default value is selected on the basis of the relative magnitude of the calculated and default values the stability, season, and time of day The larger of the two is selected for the meteorological recording station for the given hour A multiple linear regression technique is then used to provide a smooth spatial variation in mixing layer depth across the model domain

Stability classes were calculated separately for the RFP and Denver Stapleton Airport meteorological recording stations using the general classification scheme discussed in Pasquill (1961), Gifford (1961) and Turner (1964) This typing scheme employs seven stability categories ranging from A (extremely unstable) to G (extremely stable) and requires estimates of sky cover and ceiling height Cloud cover and ceiling height data for both stations were assumed to be the same and were obtained from the Denver Stapleton Airport data

Hourly average wind speed and direction also were calculated from the raw RFP meteorological data using the protocol described in EPA (1987c) An arithmetic average of the wind direction was computed first and it was then segregated into 1 of 36 10 degree increments as required by RATCHET The average wind speed for the hour was computed by taking the average of the four 15 minute data segments Hourly precipitation records from

Denver Stapleton Airport were assumed to be consistent over the entire model domain and were segregated into integer values as required by RATCHET (see Table 6)

Atmospheric Transport Model Parameters

This section describes the input parameters that were selected for the RATCHET model for simulations involving normal operational releases. These parameters include surface roughness length, topography, dry and wet deposition, diffusion coefficients, release parameters (location and height of release), and model control parameters (number of puffs per hour and computational options)

Surface Roughness Length

Roughness elements such as trees and buildings and small-scale topographic features, such as rolling hills, have a frictional effect on the wind speed nearest the surface The height and spacing of these elements will determine the frictional effects on the wind. These effects are directly related to transport and diffusion and affect atmospheric stability, wind profiles, diffusion coefficients, and the mixing-layer depth. The surface roughness length parameter is used to describe these roughness elements and is a characteristic length associated with surface roughness elements (Table 5) In RATCHET, estimates of the surface roughness length are defined for each node on the environmental grid (Figure 3) In our simulations, we selected a value of 0 6 m to represent residential and urban environs Farmland, which is predominate in the northeast part of the model domain was assigned a value of 0 05 m Range and open land consisting of rolling grass hills were assigned a value of 0.07 m Nodes that encompass the range and farmland designation were selected based on the topographic contours and land use maps The foothills and downtown Denver were assigned a value of 2 0 m and open water (Standley Lake) was assigned a value of 0 001 m

Table 5 Typical Surface Roughness Lengths for Different Land Use, Vegetation, and Topographic Characteristics (Stull 1988, Figure 9.6)

Land Use, Vegetation, and Topographic Characteristics	Surface roughness length, z_o (m)
Level grass plain	0 007–0 02
Farmland	0 02–0 1
Uncut grass, airport runways	0 02
Many trees/hedges, a few buildings	0 1–0 5
Average, North America	0 15
Average, U S Plains	0 5
Dense forest	0.3–0 6
Small towns/cities	0 6–2 5
Very hilly/mountainous regions	1 5+

Topography

The RATCHET model does not explicitly address terrain differences within the model domain Instead, topography and topographic effects on transport and diffusion are reflected in the surface roughness lengths and observed wind velocity data that are affected by topographical features Topography in the model domain (Figure 3) can be characterized by

three major features the north-south trending Colorado front range foothills in the western part of the model domain, the southwest to northeast trending Platte River Valley located in the southeast part of the model domain, and rolling hills and flat farmland that is predominate in the central and northeastern part of the model domain The surface roughness lengths reflect these features as stated in the previous section Observed meteorological data are lacking in most of the model domain and are woefully inadequate to characterize wind fields in the foothills region However, meteorological observations at Denver Stapleton Airport do capture the air movement within the Platte River Valley, which is noticeably different than that at the RFP (DOE 1980) Therefore, to a limited extent, topography is accounted in the model simulation The use of a complex terrain model would also suffer from the lack of meteorological data, especially in the foothills region This region may be of little importance because the few receptors were present there during operation of the plant

Dry and Wet Deposition

The rate of deposition of small particles on surfaces in the absence of precipitation is proportional to the concentration of material near the surface The proportionality constant between the concentration in air and the flux to the ground surface is the dry depositional velocity The current generation of applied models estimates deposition using an analogy with electrical systems as described by Seinfeld (1986) The deposition is assumed to be controlled by a network of resistances, and the depositional velocity is the inverse of the total resistance Resistances are associated with atmospheric conditions, physical characteristics of the material and the physical chemical and biological properties of the surface The total resistance in RATCHET is made up of three components aerodynamic resistance, surface-layer resistance and transfer resistance Thus, the dry deposition velocity (v_d, m s^{-1}) is calculated using

$$v_d = (r_s + r_a + r_t)^{-1} \quad (2)$$

where

r_s = surface layer resistance (s m^{-1})
r_a = aerodynamic resistance (s m^{-1})
r_t = transfer resistance (s m^{-1})

Surface layer resistance and aerodynamic resistance are given by

$$r_a = U(z)/u_*^2 \quad (3)$$

$$r_s = 2\,6/(0\,4\,u_*) \quad (4)$$

respectively where u_* = frictional velocity (m s^{-1}) and $U(z)$ = wind speed (m s^{-1}) measured at height z (m) above the ground The frictional velocity is given by

$$u = \frac{U(z)\,k}{\ln(z/z_o) - \psi(z/L)} \quad (5)$$

where k = the von Karman constant (0 4) z_o = surface roughness length ψ = stability correction factor and L = the Monin Obukhov length (m) As the windspeed increases r_s and

r_a become small resulting in unreasonably high deposition velocities The transfer resistance is used to put an upper bound on the deposition velocity The transfer resistance is the inverse of the highest measured depositional velocity for a given particle size For small particles (<1 0 μm), a transfer resistance of 100 s m^{-1} is suggested in RATCHET, and it results in calculated deposition velocities that are consistent with measured data. Measured deposition velocities over a grass surface for 0.3-μm particles and 5-m s^{-1} wind speed have been estimated to range from 2 0 × 10^{-3} to 1 8 cm s^{-1} (Harper et al 1995)

Gravitational settling (v_t) is not included in Equation (2) but may be added However, for small particles (–1 0 μm), gravitational settling is negligible compared to r_a and r_s Stokes law gives the gravitational settling velocity for particles less than 20 μm as

$$v_t = \frac{C_c\, d^2\, g\rho}{18\mu_{air}} \qquad (6)$$

where

C_c = the Cunningham Slip correction factor (dimensionless)
d = particle diameter (cm)
g = gravitational acceleration constant (980 cm s^{-2})
μ_{air} = dynamic viscosity of air (1 78 × 10^{-4} g s^{-1} cm^{-2})
ρ = particle density (1 85 g cm^{-3} for beryllium)

For particle sizes less than several microns, the Cunningham Slip correction factor is approximately 1 0 Figure 4 presents gravitational settling velocity as a function of particle size Effluent containing beryllium was reported to pass through HEPA filtration resulting in release of particle less than 1 μm in diameter Median particle size for plutonium effluent subject to the same HEPA filtration has been estimated to be 0 3 μm (Voillequé, 1997) Whicker and Schultz (1982) report gravitational settling velocities for particles less than 1 μm are insignificant compared to the other components of deposition. Depositional velocities calculated using Equation (2) ranged from 0.3 to 1 0 cm s^{-1}, for wind speeds ranging from 2 5 to 20 m s^{-1}, roughness lengths from 0 001 to 2 m, and a transfer resistance of 100 s m^{-1} Note that the gravitational settling velocity for 0 3 μm particles (=0 001 cm s^{-1}) is insignificant compared to the depositional velocity calculated with Equation (2) For our simulations, gravitational settling was ignored and a transfer resistance of 100 s^{-1} m was used

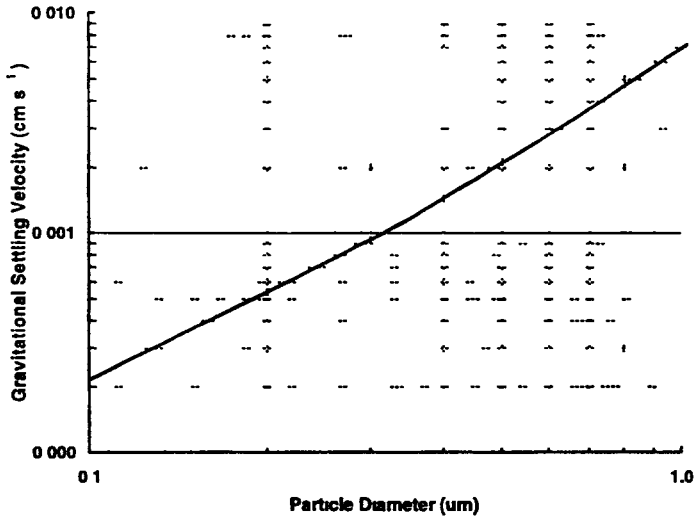

Figure 4 Gravitational settling velocity as a function of particle diameter for beryllium (ρ = 1 85 g cm^{-3})

Wet deposition of small particles in RATCHET is modeled using a washout coefficient and assuming irreversible collection of particles as the precipitation falls through the puffs The following expression discussed in Slinn (1984) is used to compute the washout coefficient in RATCHET

$$\Lambda = \frac{C\,E\,P_r}{0\,35\,P_r\,P_n^{1/4}} \tag{7}$$

where

Λ = washout coefficient (hr^{-1})
C = empirical constant assumed to have a value of 0 5
E = average collision efficiency assumed to be 1 0
P_r = precipitation rate (mm hr^{-1})
P_n = normalized precipitation rate (P_r)/ [1 mm hr^{-1}])

During periods of snow, the washout coefficient for particles is computed by

$$\Lambda = 0\,2\,P_r \tag{8}$$

Precipitation rates in RATCHET are separated into six classes three for liquid and three for frozen precipitation (Table 6) These classes are the same as reported by most airport meteorological recording stations

Diffusion Coefficients

The RATCHET model estimates the diffusion coefficients directly from statistics for atmospheric turbulence In most cases the statistics describing atmospheric turbulence (i e , standard deviation of the horizontal and vertical wind direction fluctuations) are not routinely measured at most meteorological recording stations However RATCHET makes use of atmospheric conditions that are either measured or calculated from routine meteorological data to estimate the turbulence statistics The parameters wind speed

atmospheric stability, and surface roughness are used to estimate the turbulence statistics The general form of the equation used in RATCHET for estimating the horizontal diffusion coefficient (σ_r), for the first hour following release is

$$\sigma_r = 0.5\sigma_v t \quad (9)$$

where σ_v = crosswind component of turbulence (m s^{-1}) and t is the travel time After the first hour, the horizontal diffusion coefficient is given by $\sigma_r = c_{sy}$ t where c_{sy} is a proportionality constant with dimensions of meters per second Gifford (1983) has shown the value of c_{sy} distributed between 0 14 to 1 4 with a median value of 0 5 For our simulations, we used the median value of 0 5

Table 6 Precipitation Rates and Washout Coefficients Used in RATCHET

Precipitation Type	Precipitation rate (mm hr^{-1})	Washout coefficient (hr^{-1})
Light Rain	0 1	0.254
Moderate Rain	3 0	3.26
Heavy Rain	5 0	4 78
Light Snow	0 03	0 006
Moderate Snow	1 5	0 3
Heavy Snow	3 3	0 66

The general form of the equation for estimating the vertical diffusion coefficient (σ_z), near the source is

$$\sigma_z = \sigma_w t f_z(t) \quad (10)$$

where

σ_w = standard deviation of the vertical component of the wind (m s^{-1})
$f_z(t)$ = nondimensional function related to the travel time and turbulence time scale

As a practical matter, diffusion coefficients in RATCHET are calculated in increments to avoid problems associated with spatial and temporal changes in conditions

Source Characterization

Estimated releases of beryllium to the atmosphere were provided by ChemRisk (1994a) and are summarized in a previous section Twenty-five percent of the beryllium released to the atmosphere was attributed to Building 444 and 19% was attributed to Building 776 (ChemRisk 1992) Building 444 contained the beryllium foundry where machining, casting, and milling of beryllium occurred Beryllium milling and machining did not occur in Building 776, but some materials containing beryllium were processed Therefore, beryllium was monitored in the plenum exhaust Plenum exhaust was passed through HEPA filtration before being released to the atmosphere The remaining 54% of the atmospheric beryllium releases originated from 11 other buildings surrounding Building 444 and Building 776 In Phase I, ChemRisk modeled the combined release from all buildings using a virtual stack located approximately in the center of the plant We have modeled the combined releases to originate from two points Building 776 and Building 444 Combined releases were

proportioned between the two buildings based on the relative contribution each building had to their combined total Therefore, the proportion from Building 444 was 0 25/(0 25 + 0 19) ≈ 0 6 or 60% and the remainder (40%) was proportioned to Building 776 (Table 7) Releases from Building 776 were reported to originate from five roof vents The roof vents were hooked shaped and directed flow down toward the top of the roof Therefore, the modeled release height was the height of the building The building height was 11 6 m and the horizontal dimensions were 61 m × 104 m The vents were assumed to be distributed across the roof resulting in an area source geometry The area source was simulated by modifying the initial diffusion coefficients using a procedure described by Petersen and Lavdas (1986) The initial horizontal diffusion coefficient (σ_r) is the horizontal dimension of the source divided by 4 3, and the initial vertical diffusion coefficient (σ_v) is the height of the source divided by 2 15 For these simulations, we used the 61-m length as the horizontal source dimension

Atmospheric releases of beryllium from Building 444 originated from vent number 122 after passing through two stages of HEPA filtration (ChemRisk 1992) Flow rates, stack heights, and release velocities were not characterized by ChemRisk For this analysis, we have assumed the release to occur from a point source on the roof (14 8 ft) with no buoyant or momentum driven plume rise

Table 7 Release Parameters for Building 776 and Building 444

Release Point	Parameter	Value
Building 444	Stack height	5 m
	Stack diameter	2 0 m
	Flow rate	1 6 $m^3 s^{-1}$
	UTM east	482372 m
	UTM north	4414850 m
Building 776 Roof Vents	Release height	11 6 m
	Initial σ_r	14 1 m
	Initial σ_v	5 4 m
	UTM East	482938 m
	UTM North	4415879 m

Stack tip downwash is also modeled in RATCHET, however building wake is ignored Building wake is only important for receptors close to the source At distances of about 1 km modeled atmospheric concentrations are relatively unaffected by building wakes (Ramsdell 1990)

Other Parameters

Several other parameters in RATCHET influence the accuracy of output and computer runtime These parameters include the number of puffs per hour, minimum time step, puff consolidation maximum puff radius, and minimum puff concentration at center We chose the suggested RATCHET default values for all these parameters except minimum time step and minimum concentration at puff centers (Table 8) Accuracy of the simulation can be improved by using a smaller time step The RATCHET default was 20 minutes which we reduced to 10 minutes The minimum concentration at puff centers was reduced from 1 × 10^{13} to 1 × 10^{-15} to allow for plume tracking throughout the model domain The puff

consolidation parameter value combines puffs from the same source when ratio of the puff centers to the average σ_r is less than the user-input value The puff consolidation ratio and maximum puff radius (in units of σ_r) were set at RATCHET default values of 1.5 and 3 72 respectively

Table 8 RATCHET Model Control Parameters

Model Parameter	Value
Number of Puffs per Hour	4
Minimum Time Step	10 minutes
Puff Consolidation	1.5
Maximum Puff Radius (in units of σ_r)	3.72
Minimum Concentration at Puff Centers	1×10^{-15}

Prediction Uncertainty

Prediction of annual average concentrations using a surrogate meteorological data set presents two components of uncertainty to consider First, there is the uncertainty associated with the fixed, but known quantity of the annual average concentration for a specific year at a given receptor location. For example, suppose we select a location in the model domain and measure the concentration of tracer released from the site for an entire year Let us assume the uncertainty associated with the measurement is small and inconsequential. Using the meteorological data recorded for that year, we calculate a concentration at the same receptor using an appropriate atmospheric dispersion model. Assuming our model adequately represents the physical process and phenomena (i.e , if we had the correct inputs to the model, the output would match the observations), the uncertainty associated with the model prediction results from a lack of knowledge about the correct inputs to our models Propagation of these of uncertainties through the model calculation provides a distribution of model output This is termed parameter uncertainty The output distribution may be compared with measured data to see if model predictions encompass the measurements Generally, agreement between predictions and observations is achieved when the model is adequate in representing the processes it attempts to simulate and choices regarding input parameter values have been made correctly

Model uncertainty arises from the fact that perfect models cannot be constructed, and models often fail to adequately represent the physical process they attempt to simulate. In atmospheric dispersion models, the advection-dispersion process is often oversimplified and meteorological data required to characterize turbulence in the environment are lacking In our previous example, the parameter uncertainty may not account for all differences between model predictions with observations if our model does not perfectly represent the physical process Field validation exercises provides some information as to how well a model may perform However, these are only partially relevant because field tests are generally not conducted under the same conditions that actual releases occurred

The RATCHET model has incorporated modules to explicitly assess parameter uncertainty These parameters include wind direction, wind speed, atmospheric stability class, Monin-Obukhov length, precipitation rate, and mixing-layer depths Other parameters may be assessed by simply varying the input according to some predefined distribution and

repeating the simulation a number of times until an adequate output distribution is achieved These methods are both time consuming and computationally intensive and fail to capture model uncertainty In our approach, we will ignore the built in parameter uncertainty in RATCHET and focus our efforts on defining the distribution of a correction factor that will be applied to model output The correction factor will be based on field experiments, considering the relevance of the experiment to actual release conditions and model domain environs The correction factor may then be multiplied by the model output, resulting in a distribution of concentrations at a given receptor

At this point, we have only identified the uncertainty in a predicted annual average concentration for a specific year for which we have the meteorological data for the year we are predicting Unfortunately, this is not situation we are presented with in this analysis In this analysis, we are using 5-years of meteorological data spanning a different time period (1989–1993) to define an annual average X/Q value (concentration divided by release rate) that will be applied to all previous years (1952–1988) The question is, how well does this 5-year period represent the past? We diverge from our initial example because the annual average X/Q for a known year may be considered a fixed quantity, but the annual average X/Q for *any* given year may be considered a stochastic quantity

Comparisons of annual average X/Q values computed with a 5-year data set to the annual average X/Q computed using the meteorological data for each specific year was recently performed for the Fernald Dosimetry Reconstruction Project (*RAC* 1996) Meteorological data from the Cincinnati Airport from 1987 to 1991 comprised the 5-year composite meteorological data set Annual average X/Q values computed with these data were then compared with the annual average X/Q value computed for each specific year using the meteorological data for that specific year The years spanned from 1951 to 1991 Concentrations were calculated at 160 receptors ranging in distance from 1,000 to 10,000 m from the release point A straight line Gaussian plume model for a 10 m release height was used to generate the X/Q values The 5-year composite X/Q divided by the X/Q for the specific year (P/O ratio) forms the basis of Figure 5 Figure 5 depicts the 5th, 50th and 95th percentile of the Cumulative Frequency Distribution for all points in the model domain

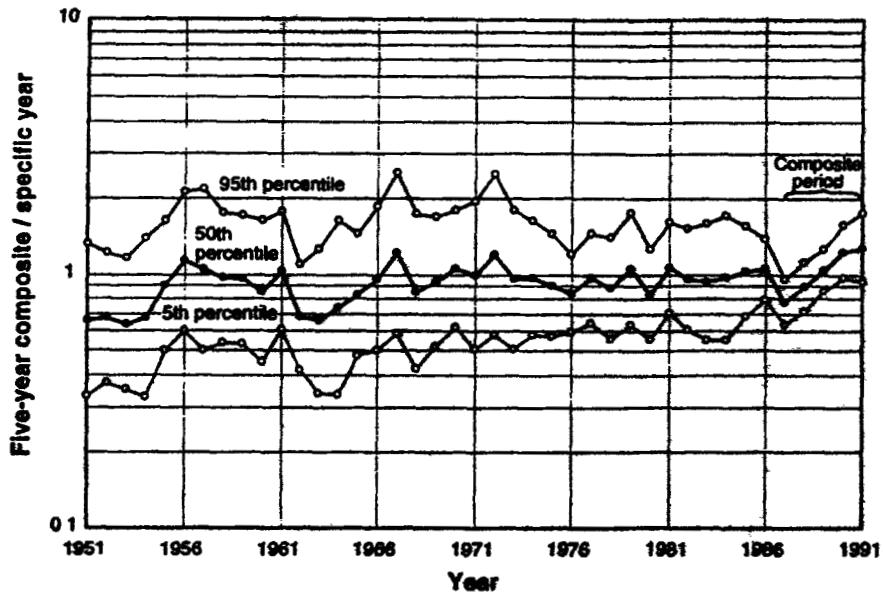

Figure 5 Distributions of P/O ratios for X/Q calculated with the Cincinnati Joint Frequency data. Predicted (P) corresponds to the five-year composite Joint Frequency for 1987–1991, observed (O) corresponds to the Joint Frequency for a specific year (from *RAC* 1996).

As one would expect, the spread is much larger for those years that do not include the 5-year composite data The long-term trend of these data may not depend strongly on location If this procedure is applied to the RFP environs using Denver Stapleton Airport data for instance, the locus of the 50th percentiles is likely to look somewhat different, although the amplitudes may be similar Obtaining these data for Denver Stapleton and performing the calculations is not a trivial task, and the overall impact on the results may not warrant the time and resource investment For this reason, we have chosen instead to adapt these data to our analysis because impacts from normal operations are not expected to dominate the overall exposures to the population surrounding the RFP and resources would be better spent on those sources that would have the greatest impact

The Fernald data were represented by a multiplicative correction factor having a geometric mean (GM) of 1 0 and geometric standard deviation (GSD) of 1 7 This distribution was developed using the following sampling scheme

1 Noting from Figure 5 that the maximum range in the GMs is a factor of two, a GM was randomly selected from a log-uniform distribution with a minimum $2^{-1/2}$ and maximum $2^{1/2}$

2 Using the GM from step (1) and GSD = 1 61 (the maximum GSD calculated from the ratio of the 5-year composite X/Q to specific year X/Q for the 40 years of data), a sample is drawn from a lognormal distribution with these parameters.

3 Values are stored from step (2) and the process is repeated

This somewhat conservative procedure takes account of the year-to-year variability in the GM of the 5-year composite X/Q to specific year X/Q ratio, as well as the uncertainty

associated with location For a sample size of 1,000, a lognormal distribution was fitted with a GM = 1 0 and GSD = 1 7

Distribution of Correction Factor We begin the process of defining the distribution of the correction factor by reviewing some field studies considered relevant to the assessment question (Table 9) The correction factor is defined for this study as the inverse of the distribution of predicted-to-observed ratio [$1/(C_p/C_o)$] The assessment question is, "What is the annual average concentration a receptor is exposed to in the model domain?" Relevant field studies included a model evaluation using the Rocky Flats Winter Validation Tracer Study data set (Rood 1996) validation exercises for RATCHET performed at the Hanford Reservation (Ramsdell et al 1994), summaries of model validations performed for the Gaussian plume model (Miller and Hively 1997), and other studies reported in the literature No one study is entirely relevant Averaging times, release conditions, meteorological conditions, and terrain conditions are different than what we are attempting to simulate in this study Nevertheless, these are the only data we have to work with and it is unlikely we will find a field validation experiment that was conducted under the exact conditions of past releases at Rocky Flats Uncertainty bounds may be expanded to compensate for our lack of knowledge

An additional study (Carhart et al 1989) not reported in Table 9 included puff dispersion models that were similar to RATCHET (MESOPUFF, MESOPLUME) Evaluations were performed using tracer data bases from Oklahoma and the Savannah River Site Oklahoma data consisted of two experiments measured at 100 and 600-km arcs downwind of a 3-hour perfluorocarbon release The Savannah River data involved 15 separate experiments 2 to 5 days in duration where ^{85}Kr was released from a 61-m stack and measures at points 28 to 144-km downwind from the source The ratio of the *average* predicted concentration to the *average* observed concentration was between 0 5 and 2 Note that this measure is different than the distribution of individual predicted-to-observed ratios reported in Table 9 There was also a tendency for models to overpredict concentrations in both data sets

The study considered most relevant to the assessment question involved the RATCHET model using the Winter Validation Tracer Study data set While it is true the release conditions for this study differed from those modeled (i e , point source and area source) and the averaging time differed (i e annual average as opposed to 9-hr average) these data were obtained in the same environs that we are attempting to simulate In addition, impacts on predicted and observed concentrations because of specific release conditions tend to diminish with increasing receptor distance Release heights are not that much different from the Winter Validation Tracer Study in which the tracer was released at 10 m above ground level Abbott and Rood (1996) have also shown that the difference between a point and a 100-m diameter area source (represented by a series of point sources distributed in a circular area) released from a height of 0–19 m is at most 5% along the plume centerline at a distance of 2 km or greater We conclude that the major difference between the Winter Validation Data set and our current situation resides with the averaging time

The largest range of predicted to-observed ratios reported in Table 9 involved complex terrain which suggests models are more sensitive to the local meteorological and terrain conditions than other factors such as release height For example note the GSD for Gaussian plume model at a highly instrumented site for ground level and elevated sources increases by

Table 9 GM and GSD of Predicted-to-Observed Ratios for Field Studies Relevant to Defining the Correction Factor for Annual Average Concentrations.

Model	Averaging Time	Receptor Distance	Release Height	Environment	GM	GSD	Comments
RATCHET[a]	9-hr	8 km	10 m	complex terrain	0 81	4.9	Rocky Flats Winter Validation Study
RATCHET[a]	9 hr	16 km	10 m	complex terrain	0 84	4 8	Rocky Flats Winter Validation Study
RATCHET[b]	28-day	20-80 km	61 m	flat	1 4	2.2	conducted at the Hanford Reservation
Gaussian Plume[c]	short-term	10 km	ground level	flat - highly instrumented		1 1	P/O ratios ranged from 0.8 to 1.2
Gaussian Plume[c]	short-term	10 km	elevated	flat highly instrumented		1.2	P/O ratios ranged from 0 65 to 1 4
Gaussian Plume[c]	short term	—	—	complex terrain		14	P/O ratios ranged from 0.01 to 100
Gaussian Plume[c]	annual average	—	—	complex terrain		3 8	P/O ratios ranged from 0 1 to 10
Gaussian Plume[c]	annual average	10 km	ground level	flat		1.5	P/O ratios ranged from 0.5 to 2
Gaussian Plume[c]	annual average	10 to 150 km	ground-level	flat		2.2	P/O Ratios ranged from 0.25 to 4
Gaussian Plume[d]	12 hr	1 to 5 km	60 m	relatively flat	0 82	3 4	terrain heights varied by about 50 m
Gaussian Plume[d]	72 hr	1 to 5 km	60 m	relatively flat	0 67	2.1	terrain heights varied by about 50 m
Eulerian and Gaussian Plume[e]	annual average	1 to 1,000 km	0 to 60 m	relatively flat	0.75	1 5	Gaussian model used for receptors out to 50 km
CTDMPLUS[f]	12 to 72 hr	1 km	—	complex terrain	1 6	2.5	EPA complex terrain model

a Rood 1996
b Ramsdell et al. 1994
c Miller and Hively 1987
d Robertson and Berry 1989
e Simpson et al. 1990
f Genikhovich and Schiermeier 1995

about 9 % while the difference between the GSD for flat and complex terrain is almost an order of magnitude It is also evident from these data that uncertainty decreases with increasing averaging time (Miller and Hively 1987) and uncertainty also tends to increase with increasing receptor distance (>10 km), at least for the Gaussian plume model.

One factor not considered in any of the field studies was deposition and plume depletion Most field studies use inert tracers to avoid additional complications involving plume depletion and deposition However, Miller et al. (1978) illustrates that deposition and plume

depletion have little impact on inhalation dose for deposition velocities less than1 0 cm s^{-1} and release heights greater than 50 m for receptors within 10 km of the release point For ground level releases plume depletion has a greater effect The ratio of the depleted to non depleted plume was 0 02 for deposition velocities in the 1 0 cm s^{-1} range and 0 67 for deposition velocities in the 0 1 cm s^{-1} range Beryllium was not released at ground level or at 50 m and deposition velocities ranged from 0 3 to 1 0 cm s^{-1} Therefore, the actual amount of plume depletion is somewhere between these values Deposition velocities in the 1 0 cm s^{-1} range are associated with roughness lengths of around 2 0 m which are limited to the foothills region of the model domain where few receptors are present For these reasons, we have not treated the uncertainty associated with deposition velocities and plume depletion explicitly

With the distribution of predicted-to-observed ratios for RATCHET from the Winter Validation Tracer Study as our starting point, our approach is to modify this distribution based on the differences between the study conditions and those of past releases, and our assessment question We combined data points at the 8 and 16 km distance into a composite set and justified this action based on the evaluations in Rood (1996) that showed similar GM and GSD values for 8 and 16-km data In addition, the confidence intervals on the geometric

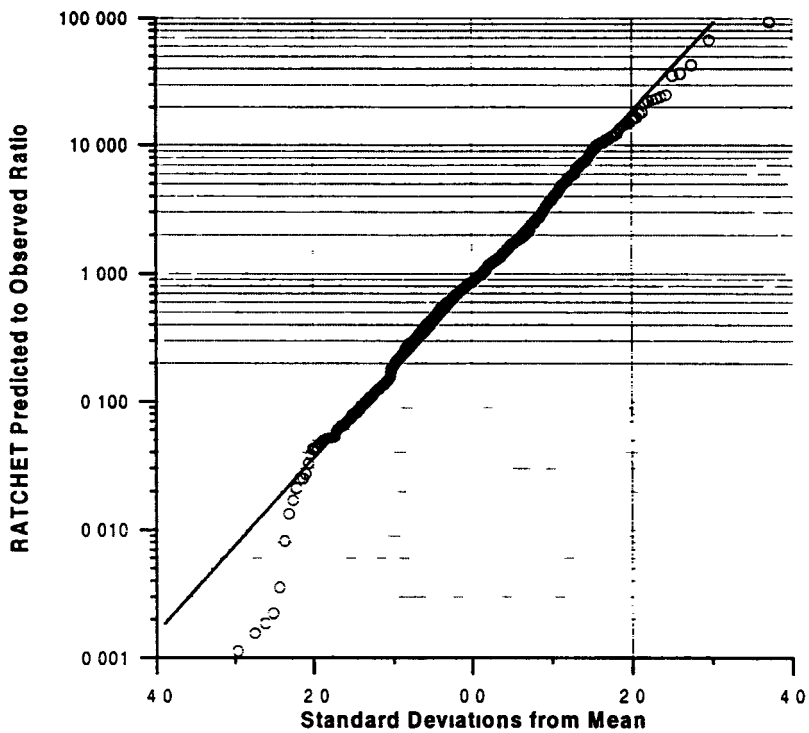

Figure 6 Predicted to-observed ratios for the RATCHET model as a function of standard deviation from the mean (normalized to a mean of 0 and standard deviation of 1) The solid line represents the lognormal fit to the distribution Circles represent individual data points

mean and variance of the observed-to-predicted ratio overlapped. The composite distribution had a GM of 0 82 and GSD of 4 7 Predicted-observed ratios are plotted as a function of the number of standard deviations from the mean (normalized to the standard normal distribution) in Figure 6 Note that most of the data points (± 2σ) lie along the line representing the lognormal fit to the data, with the exception of the tails We, therefore, represent the distribution of predicted-to-observed ratios with a lognormal distribution with a GM and GSD as defined above Points on the tails, particularly those with predicted-to-observed ratios less than 0 01, were associated with Test 5 (February 9, 1991) at the 8-km arc in the E–NE sector for the hours 16 00 to 18 00 All models performed poorly for this test. Concentrations in E–NE sector were grossly underestimated (greater than a factor of 10 difference) and the ground-level contaminant mass at 8 km was also underestimated. Models appeared to have difficulty responding to the transition from daytime to nighttime stability conditions During the latter hours of the test and under predominately nighttime conditions (18 00–23 00), predicted concentrations showed better agreement with the observations

As stated previously, the major difference between the Winter Validation Tracer Study data and the assessment question is the averaging time Averaging time appears to have a large impact on the uncertainty bounds For example, Simpson et al. (1990) reports the GSD of the predicted-to-observed ratio is reduced 38% with an increase in averaging time from 12 to 72 hours (Table 9) Also note the GSD for the short-term and annual average predicted-to-observed ratio for the Gaussian plume model under complex terrain conditions increases from 3 8 to 14 Validation exercises performed with RATCHET at the Hanford Reservation for an elevated release at distances greater than 20 km showed a slight overprediction by the model (GM=1 4) and a GSD value of 2 2, which is smaller than the GSD for the Winter Validation Tracer Study data It is not clear whether these differences are due to averaging time, release height or terrain, or receptor distance

These field studies indicate the GSD be reduced for averaging time In addition, several studies indicate model over-prediction at distances greater than 20 km Noting that the GSD for annual average predicted-to-observed ratios ranged from 1.5 for the Gaussian plume model under flat conditions to 3 8 under complex terrain conditions, and the RATCHET model had a GSD values of 4 7 for short-term releases in complex terrain and 2 2 for monthly averages in flat terrain, we have choose a GSD value of 3 0 for our simulations We retain the GM of 0 82 for receptors within 20 km of the release point and increase this value to 0 9 for receptors at distances greater than 20 km Validation studies indicate predicted-to-observed ratios greater than 1 0 (reflecting model overprediction) at distances greater than 20 km. While this may be true, we have no site-specific data to verify this observation for our model domain The lower GM value will potentially result in model overprediction, and, thereby provide at least a conservative estimate of concentrations at these distances. The distribution was truncated by the minimum (0 001) and maximum (100) predicted-to-observed ratio calculated for RATCHET using the Winter Validation Tracer Study data.

Summary of Prediction Uncertainty. Two correction factors are applied to our model predictions One correction factor accounts for the uncertainty associated with the prediction of the annual average concentration for a specific year The other correction factor accounts for the uncertainty associated with using a 5-year composite meteorological data set (1989–1993) to predict the annual average concentration for years past (1953–1988) Correction

factors are represented by lognormal distributions The distribution for an annual average prediction for a specific year was truncated by the minimum and maximum predicted-to-observed ratio encountered while evaluating RATCHET using the Winter Validation Tracer Study data The correction factor for the annual average concentration for a specific year is given by the inverse of the distribution of the predicted-to-observed ratios, which has a GM of 0 82 and GSD of 3 for receptors within 20 km of the source and a GM of 0 9 and GSD of 3 for receptors greater than 20 km from the source The correction factor associated with using a 5-year composite meteorological data set to predict the annual average concentration for years past has a GM of 1 0 and a GSD of 1 7

Annual Average X/Q Values

The procedure and models described in the previous sections were used to calculate an annual average X/Q (concentration divided by source term [s m^{-3}]) for all concentration grid nodes in the model domain Grid node spacing for the concentration grid was set at 1,000 m Annual average X/Q values were calculated separately for releases from Building 444 (Figure 7) and Building 776 (Figure 8) The annual average concentration at each of the concentration grid nodes for each year of meteorological data (1989–1993) were computed for a constant unit release (1 mg s^{-1}) from each building The five concentration values at each grid node were then averaged to yield a 5-year composite annual average concentration

The dispersion patterns shown in Figures 7 and 8 are characterized by a large east-southeast trending component and a smaller westerly component Wind roses constructed using RFP data from 1984–1993 (DOE 1995a) indicate the predominant wind direction to be from the west northwest so the east trending plume component appears reasonable Near the Denver metropolitan area and after considerable dilution, the plume appears to move in a northeast southwest pattern We attribute this feature to the influence of air movement within the Platte River Valley and the diurnal pattern of upslope-downslope conditions that characterize the general air movement on the Colorado Front Range environs Downslope conditions typically occur during the evening hours and are characterized by drainage flow of cooler air from the foothills to the plains Westerly winds predominate, but the direction may be altered by local topography Upslope conditions are a result of daytime heating and typically result in easterly winds that prevail during the daylight hours with transition from upslope to downslope conditions occurring during the evening and transition from downslope to upslope occurring during the morning During evening hours under stable conditions, cool air near the surface drains from the Denver metropolitan area down the Platte River Valley (which flows to the northeast) and out to the plains During daylight hours and after surface heating has eliminated the cooler surface layer the downslope conditions cease This is followed by a brief period of relatively calm winds which in turn is followed by return of air up the valley or upslope conditions Meteorological data at Denver Stapleton Airport captures these transitions in the Platte River Valley that are reflected in the X/Q isopleth maps

Perhaps the most convincing subjective argument for the validity of the predicted dispersion patterns is an illustration described in "Air Pollution in the Denver Area" published by the Public Service Company of Colorado (Crow 1967) The illustration shows the typical daily airflow patterns related to air pollution in the Denver metropolitan area These dispersion patterns have a prominent westerly component along with the northeasterly

component formed by movement up and down the Platte River Valley and show the same general trend of dispersion predicted by the model simulations in this study

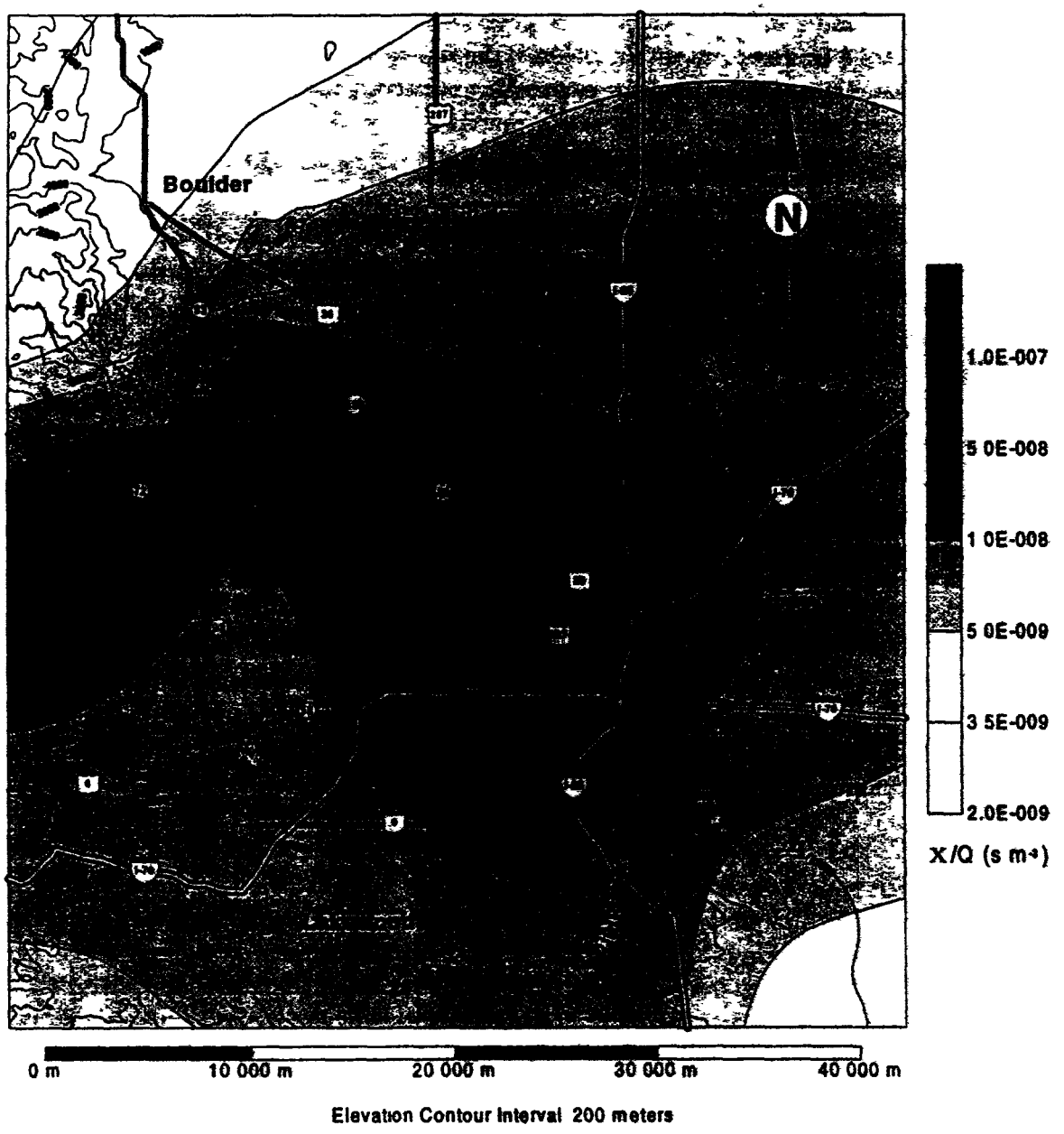

Figure 7 Isopleth map of the annual average X/Q for particulate releases from Building 776 using meteorological data from the RFP and Denver Stapleton Airport from 1989–1993

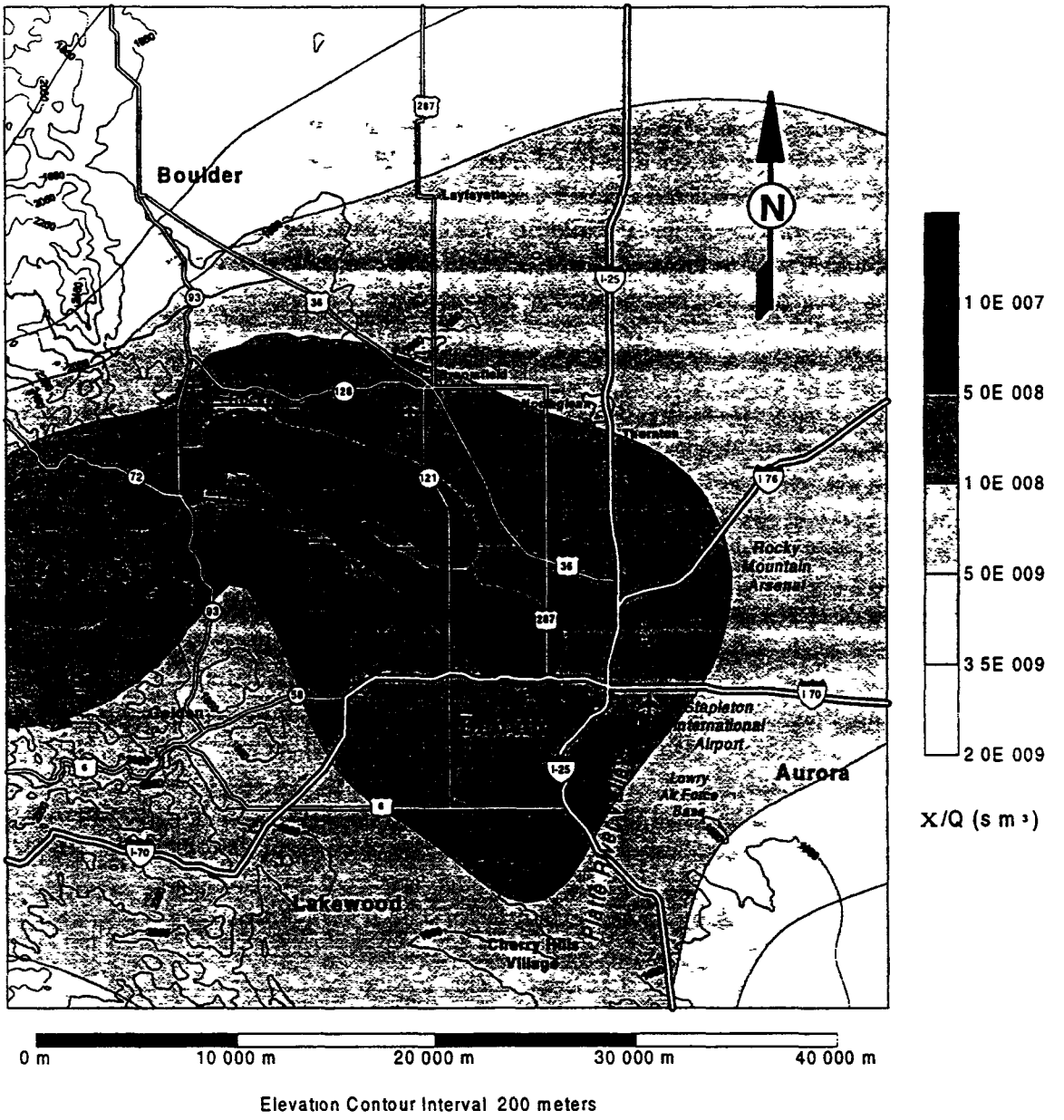

Figure 8 Isopleth map of the annual average X/Q for particulate releases from Building 444 using meteorological data from the RFP and Denver Stapleton Airport from 1989–1993

Predicted Concentrations

Predicted concentrations of beryllium at specific receptors were calculated for each year in which source term information was available Uncertainty in the predicted concentration included the uncertainty in the dispersion estimate and source term The concentration for the i^{th} year for releases from a given building (Building 444 or 776) is given by

$$C_i = \frac{\chi}{Q} Q_i \, CF_1 \, CF_2 \tag{11}$$

where Q_i = the annual release of beryllium for the i^{th} year for a given building (776 or 444), CF_1 = the correction factor for prediction of the annual average concentration for a specific year, CF_2 = uncertainty associated with using a 5-year composite meteorological data set The correction factors and Q_i are stochastic quantities Therefore, the concentration is also stochastic quantity The predicted concentration a receptor was exposed to was the sum of the prediction concentration from Building 776 and Building 444 releases For risk calculation purposes, we are interested in the integrated concentration a receptor may have been exposed to while residing in the vicinity of Rocky Flats

EXPOSURE SCENARIOS AND RISK CALCULATIONS

Calculation of risk from historical beryllium releases from RFP was limited to two hypothetical exposure scenarios The first scenario considered a male rancher who lived and worked east of the RFP along Indiana Street where the maximum concentration outside the buffer zone was predicted to occur (UTM coordinates 484320E 4415060N) The second was a female office worker who lived near the rancher, but who worked in downtown Denver near 17th Street and Lawrence (UTM coordinates 500222E 4398550N) The rancher was assumed to live and work at the same location for the entire period the RFP operated The office worker was assumed to have moved and lived in the area from 1975 to the present. As discussed earlier, inhalation was the only pathway of exposure considered in the assessment Ingestion of beryllium in water, food, and inhalation of beryllium attached to resuspended soil are potential pathways that could have been considered in more detail However, beryllium compounds are very insoluble and tend to adhere to soil and sediments Ingested beryllium is poorly absorbed and beryllium does not bioaccumulate in fish or other meat. Preliminary ingestion risk estimates determined in Phase I were calculated by multiplying a SF by the ingestion dose for one year (1968) for one location (sector 12 located southeast of RFP), then dividing the risk by an absorption factor of 0 01 (ChemRisk 1994c) Applying the absorption factor is not recommended when risk is calculated using EPA-derived SF which are designed to be used with intake concentrations, rather than absorbed dose (EPA 1995) Application of the absorption factor overestimated the risk by a factor of about 10^2 The GM of the estimated risks reported in Appendix N of the Phase I Task 8 report would be reduced from 1×10^{-12} to 1×10^{-14} if this factor were not applied

Because the risk from ingestion of beryllium is so low, and resuspension of beryllium in soil appears to have been a small contributor to overall risk, the additional time and resources required to include these pathways in the Phase II risk calculations was not thought to be warranted All risk calculations were performed using Monte Carlo sampling and the Crystal Ball software (Decisioneering 1996)

The male rancher scenario (Table 10) assumed the individual spent 8 hours per day at work, 8 hours per day doing other activities, and 8 hours per day sleeping During working hours, 7 hours were spent doing light exercise and 1 hour were spent performing heavy exercise During nonworking waking hours, 4 hours were spent sitting, 3 hours were spent doing light activity, and 1 hour was spent in heavy exercise Occupational and

nonoccupational breathing rates were adjusted according the time fraction spent doing these various activities Indoor shielding was not considered

The female office worker scenario (Table 10) assumed the individual spent 8 hours per day at the office in Denver 8 hours per day at home and 8 hours per day sleeping (commuting time was not considered) During working hours 1/3 of the time was spent sitting and 2/3 of the time in light exercise During nonworking waking hours at home, 4 hours were spent sitting, 3 hours were spent in light activity, and 1 hour was spent doing heavy exercise Occupational and nonoccupational breathing rates were adjusted according the time fraction spent doing these various activities Indoor shielding was not considered

Cancer risk from the inhalation of beryllium was calculated using the standard risk equations described in EPA (1989) and given by Equation (12)

$$R = \frac{SF\ I}{BW\ AT} \tag{12}$$

where

R = cancer risk
SF = carcinogenic slope factor (kg d mg^{-1})
I = integrated contaminant intake (mg)
BW = body weight (kg)
AT = averaging time (70 years × 365 days per year)

Integrated contaminant intake is the total amount of beryllium originating from RFP inhaled by the receptor over their lifetime Typically, this is calculated by assuming a constant concentration and multiplying by a breathing rate, exposure frequency (number of days per year exposed), and exposure duration (number of years the receptor is exposed) We used Equation (13) to arrive at integrated intake that accounts for different breathing rates and year-to-year variation in air concentration

$$I = \sum_{n}^{i=1} C_i \left(BR_1\ f_1 + BR_2 f_2 + BR_3 f_3 \right) EF \tag{13}$$

where

C_i = the annual average concentration for the i^{th} year (mg m^{-3})
$BR_{1\,2\,3}$ = the breathing rate for occupational non-occupational and resting activity respectively (m^3 d^{-1})
$f_{1\,2\,3}$ = the fraction of time spent in occupational non-occupational and resting activity respectively
EF = exposure frequency (365 days for each year exposed)
n = number of years exposed

Table 10 Parameter Values and Distributions for the Exposure Scenarios and Risk Calculation

Parameter	Value	Distribution Type	5th	50th	95th
Breathing rate male rancher occupational (m^3 d^{-1})	13 6				
Breathing rate male rancher non-occupational (m^3 d^{-1})	9 7				
Breathing rate, male rancher sleeping (m^3 d^{-1})	3 6				
Breathing rate female non-occupational (m^3 d^{-1})[a]	8				
Breathing rate, female occupational (m^3 d^{-1})[b]	7.9				
Breathing rate female, sleeping (m^3 d^{-1})	2 6				
Breathing rate correction factor	n/a	custom	0 71	1 0	1.4
Body weight (kg)	n/a	normal	45	71	97
Slope Factor (kg d mg^{-1})[c]	n/a	triangular	0.56	8.4	25

a Breathing rate at home doing indoor/outdoor activities

b Breathing rate for an office worker

c Values presented in the 5th 50th and 95th percentile are the minimum, most likely and maximum value for the triangular distribution.

For the office worker, intake was computed separately for each location (Denver office building and residence) The fraction of time spent at each activity (occupational, nonoccupational, and resting) were set to zero depending on location of the office worker For example, the *f* value for nonoccupational and resting activities was set to zero for computing intake to the office worker while working in Denver Each receptor was assumed to spend 100% of the time in their daily routine (i.e , no vacation or extend periods of time away from the Denver area) While this assumption is unrealistic, it does provide a bounding estimate of the estimated risk.

Uncertainty distributions were assigned to body weight, breathing rate, and the carcinogenic SF The remaining parameters were considered fixed. Distributions for body weight and breathing rate were obtained from Finley et al. (1994) Median estimate breathing rates for occupational, nonoccupational, and sleeping activities were estimated from breathing rate data presented in Roy and Courtay (1991) for heavy exercise, light exercise, and resting The median estimate occupational and nonoccupational breathing rate was the time-weighted average breathing rate for resting (applied to time spent sitting), light, and heavy exercise The median value breathing rate assigned to sleeping was the breathing rate for resting The median estimated breathing rates for occupational, nonoccupational and resting activities were then multiplied by a correction factor based on data in Finley et al (1994) The distribution of breathing rates for individuals 30–60 years old were represented by percentiles on the cumulative frequency distribution Each breathing rate corresponding to a given percentile on the cumulative frequency distribution was normalized to the 50th percentile value (14 8 m^3 d^{-1}) The distribution of the normalized rates were then input into Crystal Ball as a custom distribution.

Distribution of the SF was based on the risk per unit concentration (m^3 mg^{-1}) values reported in the TOMES (Micromedex Inc 1996) database which was then converted to SFs (please see discussion on SFs in a previous section) A triangular distribution with the most likely value being the arithmetic mean of the SFs (8 4 kg d mg^{-1}) was implemented

The rancher and office worker residence were located where the maximum concentration outside the buffer zone and along Indiana Street was calculated The maximum annual average X/Q at that location was $1\,2 \times 10^{-6}$ s m^{-3} for Building 776 releases and $1\,5 \times 10^{-6}$ s m^{-3} for Building 444 releases The 5th and 95th percentile of the maximum predicted beryllium concentration at that location ranged from $4\,3 \times 10^{-7}$ to $3\,4 \times 10^{-5}$ µg m^{-3} respectively and occurred in the year 1968, the year of highest releases The 5th and 95th percentile of predicted beryllium concentration at the highest concentration node in the model domain (1 km east of RFP) was $1\,8 \times 10^{-6}$ µg m^{-3} to $1\,4 \times 10^{-4}$ µg m^{-3} respectively This can be compared with a natural background range of 3×10^{-5} µg m^{-3} to 3×10^{-4} µg m^{-3}, (median of 1×10^{-4} µg m^{-3}) and monthly average onsite sampler range of 7×10^{-4} to $1\,5 \times 10^{-3}$ µg m^{-3} reported in Rope et al (1997)

RISK ESTIMATES

Median cancer risk estimates for beryllium inhalation (50th percentile) were $5\,8 \times 10^{-10}$ for the rancher and $2\,1 \times 10^{-11}$ for the office worker Estimated risks (5th and 95th percentile values) ranged from $5\,3 \times 10^{-11}$ to $6\,1 \times 10^{-9}$ for the rancher and $1\,8 \times 10^{-12}$ to $2\,3 \times 10^{-10}$ for the office worker (Table 11 and Figure 9) These estimates are well below the EPA point of departure for acceptable risks ($1 \times 10^{-4} - 1 \times 10^{-6}$) The risks presented in this report are not comparable to risks calculated in Phase I and reported in the Task 8 report (Chemrisk 1994c) Differences are summarized as follows

- Phase I reported the risk from 1 year of exposure The risks presented in this report represent integrated lifetime exposure to airborne releases of beryllium from RFP while the receptor resided in the model domain
- Risks from ingestion of contaminated food stuffs and inhalation of resuspended material that were computed for Phase I are not considered in this analysis
- Absorption factors for inhalation (0 5) and ingestion (0 01) that were used in Phase I were considered inappropriate and not used in Phase II

Table 11 Percentiles of the Cumulative Frequency Distribution for Predicted Carcinogenic Risk from the Inhalation of Beryllium

Percentile	Carcinogenic risk (Rancher)	Carcinogenic risk (Office)	Percentile	Carcinogenic risk (Rancher)	Carcinogenic risk (Office)
0	$3\,3 \times 10^{-12}$	$1\,3 \times 10^{-13}$	55	$7\,0 \times 10^{-10}$	$2\,5 \times 10^{-11}$
5	$5\,3 \times 10^{-11}$	$1\,8 \times 10^{-12}$	60	$8\,4 \times 10^{-10}$	$2\,9 \times 10^{-11}$
10	$8\,8 \times 10^{-11}$	$3\,1 \times 10^{-12}$	65	$1\,0 \times 10^{-09}$	$3\,7 \times 10^{-11}$
15	$1\,3 \times 10^{-10}$	$4\,6 \times 10^{-12}$	70	$1\,2 \times 10^{-09}$	$4\,4 \times 10^{-11}$
20	$1\,7 \times 10^{-10}$	$6\,0 \times 10^{-12}$	75	$1\,5 \times 10^{-09}$	$5\,5 \times 10^{-11}$
25	$2\,1 \times 10^{-10}$	$7\,7 \times 10^{-12}$	80	$1\,9 \times 10^{-09}$	$7\,0 \times 10^{-11}$
30	$2\,7 \times 10^{-10}$	$9\,5 \times 10^{-12}$	85	$2\,6 \times 10^{-09}$	$9\,2 \times 10^{-11}$
35	$3\,3 \times 10^{-10}$	$1\,1 \times 10^{-11}$	90	$3\,6 \times 10^{-09}$	$1\,3 \times 10^{-10}$
40	$4\,0 \times 10^{-10}$	$1\,4 \times 10^{-11}$	95	$6\,1 \times 10^{-09}$	$2\,3 \times 10^{-10}$
45	$4\,7 \times 10^{-10}$	$1\,7 \times 10^{-11}$	100	$1\,4 \times 10^{-07}$	$3\,3 \times 10^{-09}$
50	$5\,8 \times 10^{-10}$	$2\,1 \times 10^{-11}$			

The scenario involving the rancher may be considered the maximum exposed individual in the model domain because he was placed at the point of highest concentration outside the RFP buffer zone and remained there for the entire operating period of the plant. However, it is recognized that ranchers could have been grazing cattle within the current buffer zone and up to the old cattle fence There were also bunkhouses or some type of permanent overnight ranch camp to the northeast within the buffer zone To increase the risk substantially from our estimates, the concentration within the buffer zone would have to be several orders of magnitude greater than outside it This simply is not the case as is evidenced by the X/Q plots provided previously in the report and differences between the predicted concentration at Indiana Street and the maximum concentration in the model domain. The resulting risk, accounting for occupancy time while exposed to concentrations within the buffer zone, would still be at or below the EPA point of departure of 1×10^{-4} to 1×10^{-6}

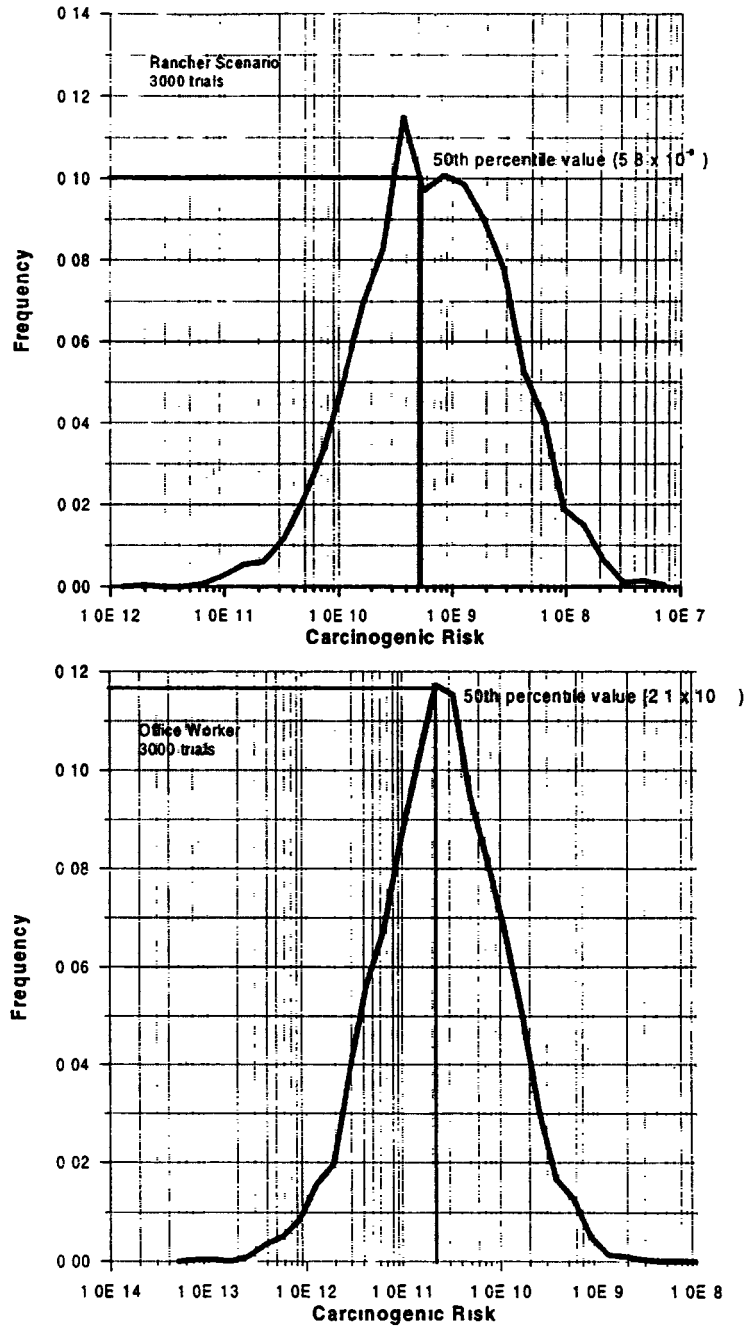

Figure 9 Frequency distributions of predicted cancer risk from inhalation of beryllium for the rancher and office worker scenario Distributions were generated with Crystal Ball simulation software

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